Quantum and Nonlinear Optical Imaging

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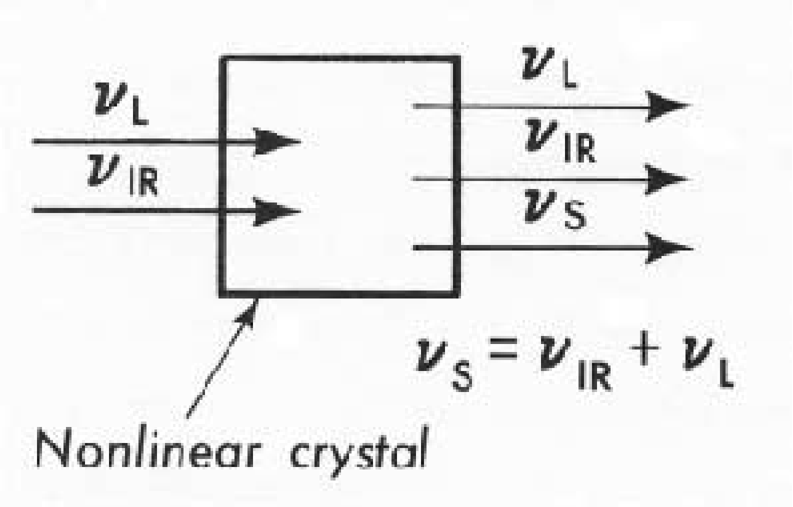
- Imaging upconversion
  (for astronomy and THz imaging)
- The promise of quantum imaging
  Quantum (?) lithography
  Quantum (?) coincidence imaging
- Generation of quantum states of light
- Development of nonlinear optical materials (enabler)
  Composite materials
  Nanofabrication
- Nonlinear optical microscopy
- Underlying issues in nonlinear optics

Presented at the ARO Optics Workshop, October 16, 2002.

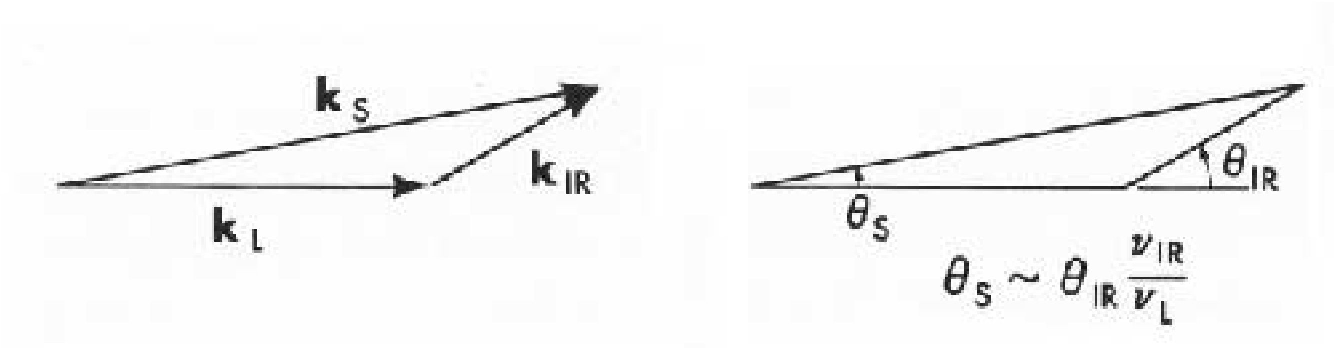
Imaging Upconversion

"Noise-free" conversion of infrared images to the visible.

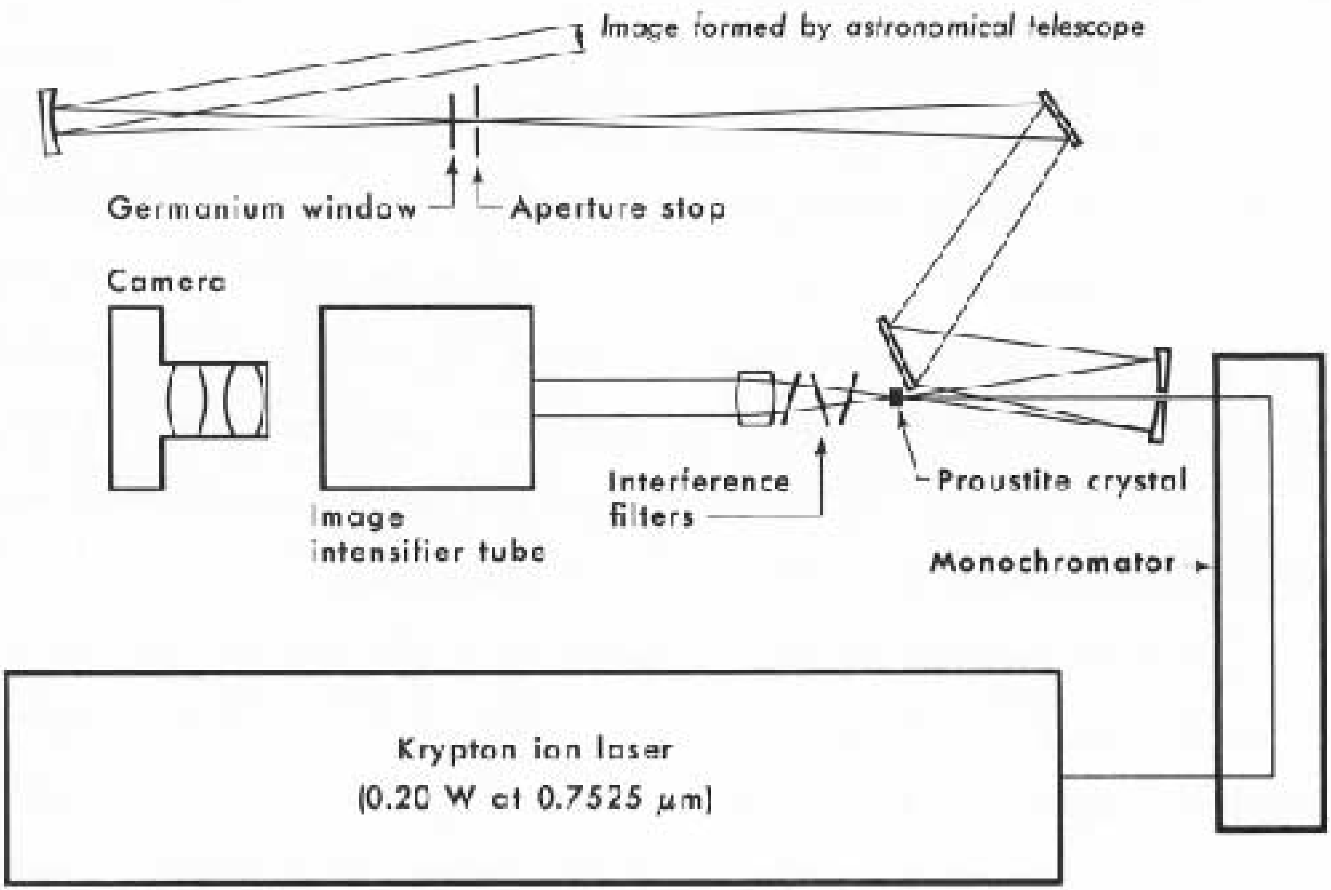
Proposed by Midwinter and Warner (1967).



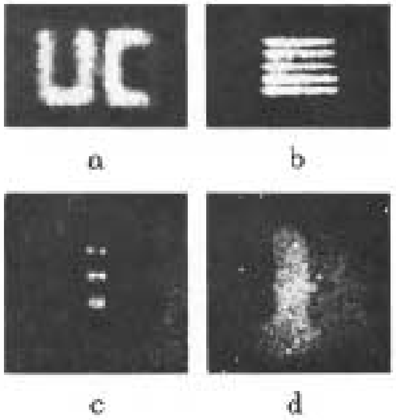
Phase-matching requirements ensure that image information is preserved.



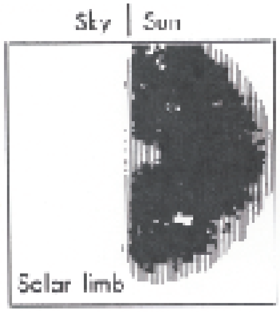
Astronomical Imaging Upconversion



laboratory sources



astronomical sources

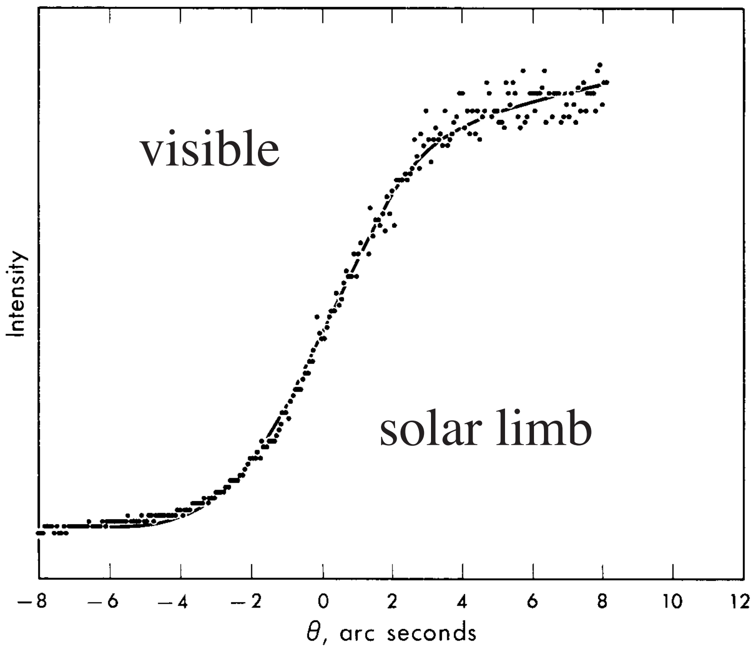


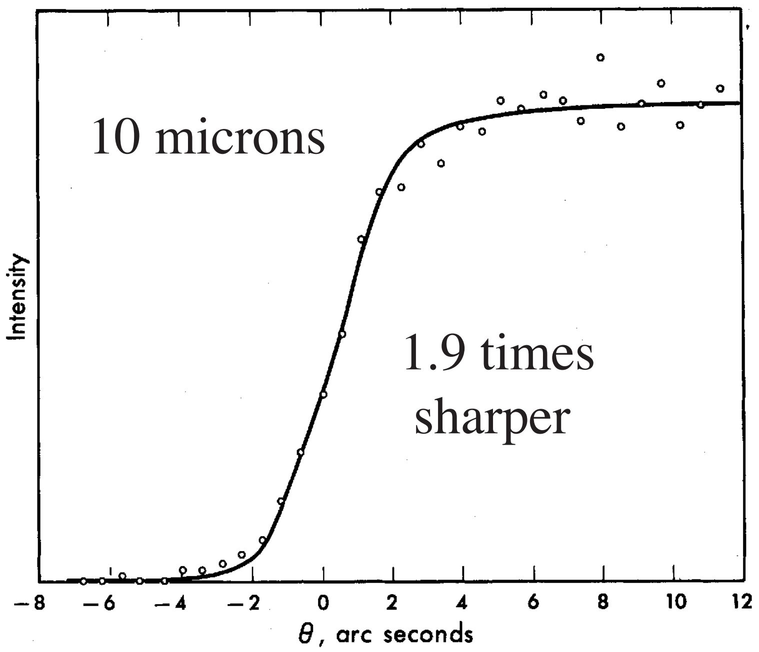
R. W. Boyd and C. H. Townes Appl. Phys. Lett. 33 440 (1977).

Resolution of Astronomical Telescopes

- Wavelength dependence under turbulence-dominated conditions
- Images are sharper in the infrared than in the visible!

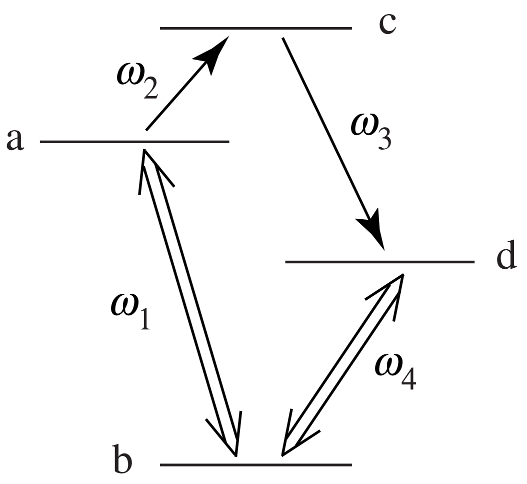
  (D. L. Fried, R. E. Hufnagel, V. I. Tatarski)
- IR data obtained using infrared upconversion



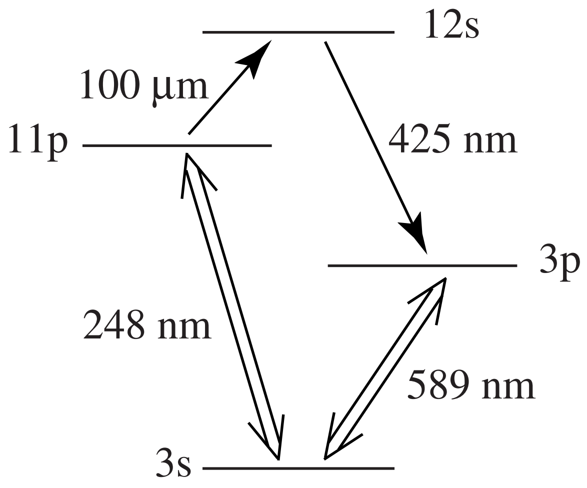


R. W. Boyd, J. Opt. Soc. Am. 68, 877, 1978.

Efficient Far IR and THz Imaging by use of EIT



Basic concept of our approach. Because of strong saturation of the lower transitions, upconversion occurs with essentially unit efficiency.



Sodium energy levels for the conversion of 100 micron radiation to the visible.

R. W. Boyd and M. O. Scully, Appl. Phys. Lett. 77, 3559, 2000.

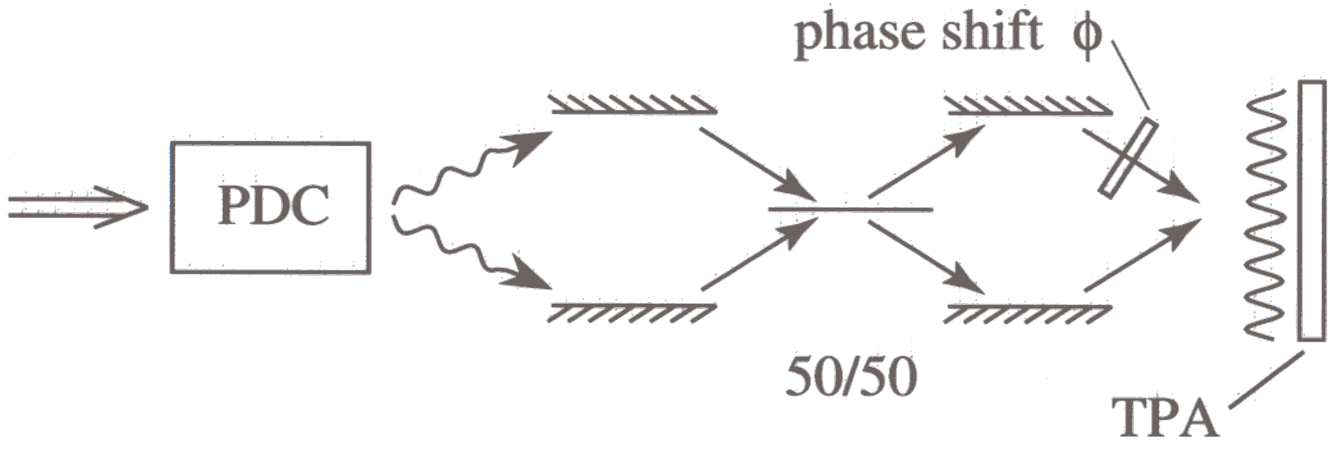
Efficient Far IR and THz Imaging by use of EIT

R W. Boyd and M. O. Scully

- EIT concepts allow “upconversion” of IR images to the visible with high quantum efficiency (approaching unit efficiency) .
- Upconversion is a “noise-free” process; only noise in output is (quantum) noise of IR signal.
- Technique holds promise of unprecedented sensitivity of FIR and THz detection (detection of single THz quanta)!
- Applications include FIR astronomy and THz imaging of biological tissue.
- Pitfall: very narrow spectral acceptance bandwidth.

Quantum Lithography and Microscopy

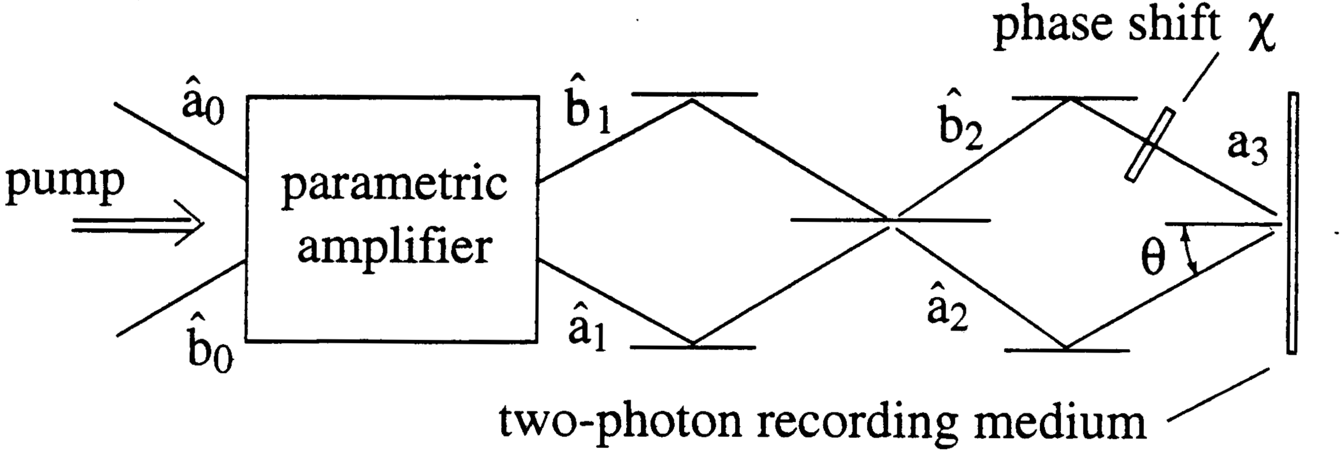
- Entangled photons can be used to form interference patterns with detail finer than the Rayleigh limit
- Process “in reverse” performs sub-Rayleigh microscopy



Boto et al, Phys. Rev. Lett. 85, 2733, 2000.

Use of High-Gain Parametric Amplifier

Is two-photon interference pattern preserved?



- Transfer equations of OPA

$$\hat{a}_1 = U\hat{a}_0 + V\hat{b}_0^\dagger, \quad \hat{b}_1 = U\hat{b}_0 + V\hat{a}_0^\dagger$$

where

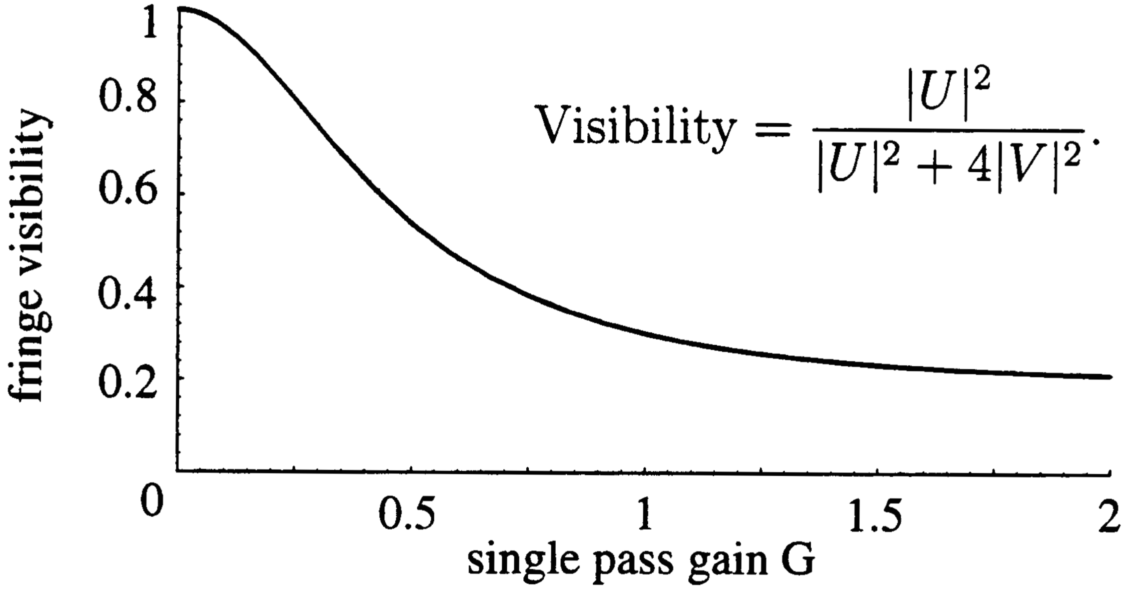
$$U = \cosh G \qquad V = -\, i \exp(i\varphi) \sinh G$$

- Field at recording medium

$$\hat{a}_3 = \frac{1}{\sqrt{2}} \left[(-\,e^{i\chi} + i)(U\hat{a}_0 + V\hat{b}_0^\dagger) + (ie^{i\chi} - 1)(U\hat{b}_0 + V\hat{a}_0^\dagger)\right]$$

- Two-photon absorption probablility

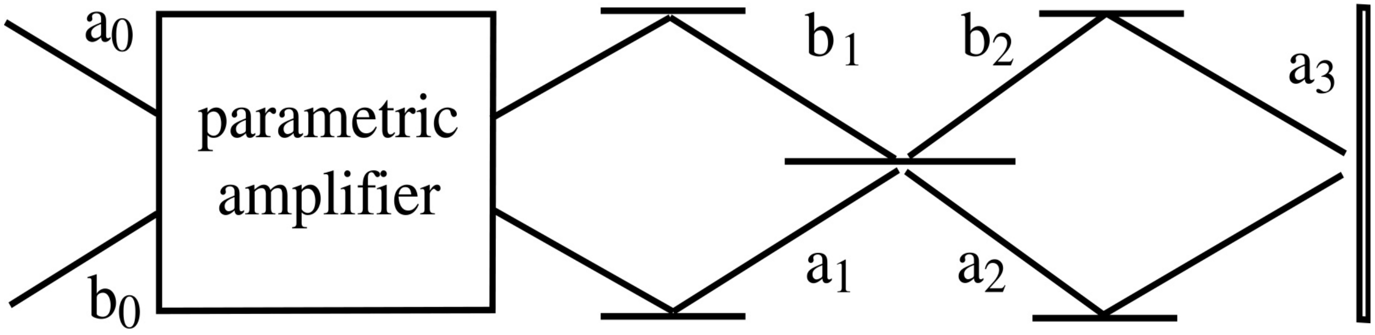
$$\langle 0,0|\hat{a}_3^\dagger \hat{a}_3^\dagger \hat{a}_3 \hat{a}_3|0,0\rangle = 4|V|^2\left[|U|^2 \cos^2\chi + 2|V|^2\right]$$



$$\text{Visibility} = \frac{|U|^2}{|U|^2 + 4|V|^2}.$$

(Phys. Rev. Lett. 86, 1389, 2001)

QUANTUM LITHOGRAPHY PROPOSAL



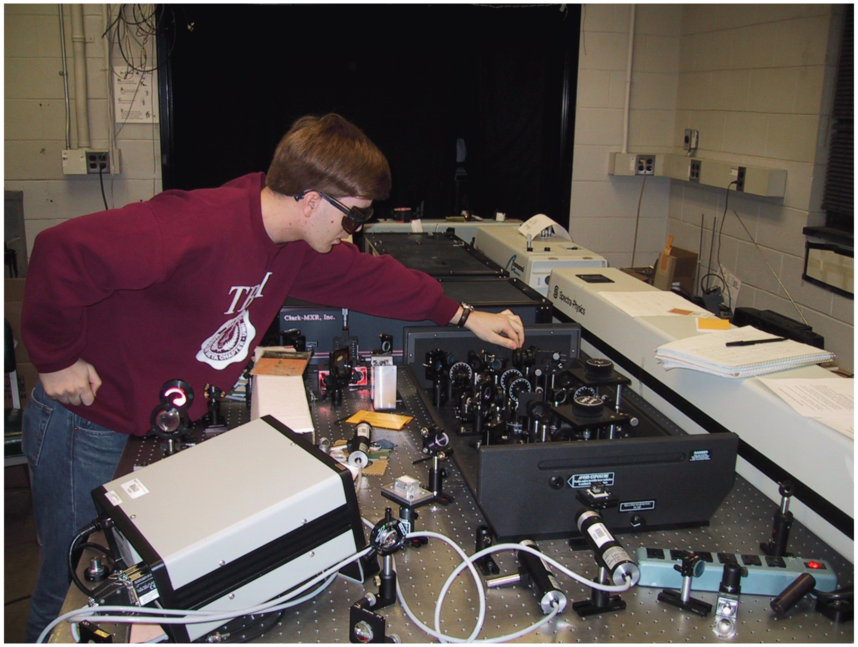
G. S. Agarwal, R. W. Boyd, E. M. Nagasako, S. J. Bentley, Phys. Rev. Lett., 86, 1389, 2001.

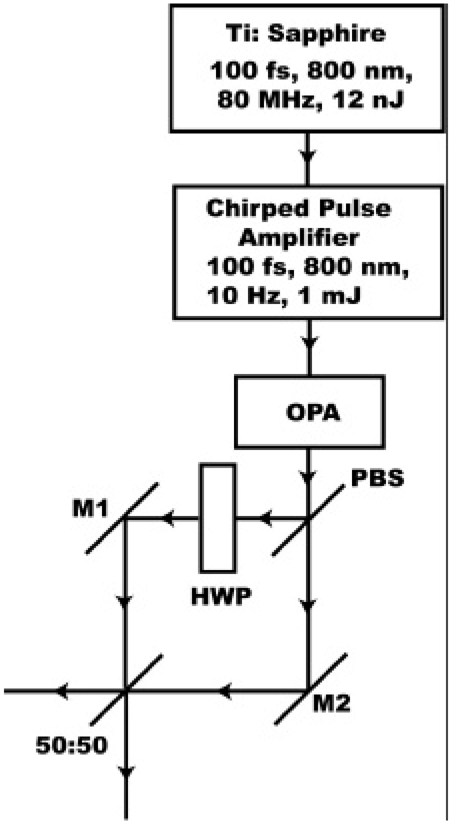
E. M. Nagasako, S. J. Bentley R. W. Boyd, and G. S. Agarwal, Phys. Rev. A, 64, 043802 (2001).

E. M. Nagasako, S. J. Bentley and R. W. Boyd, and G. S. Agarwal, J. Mod. Optics, 49, 529 2002

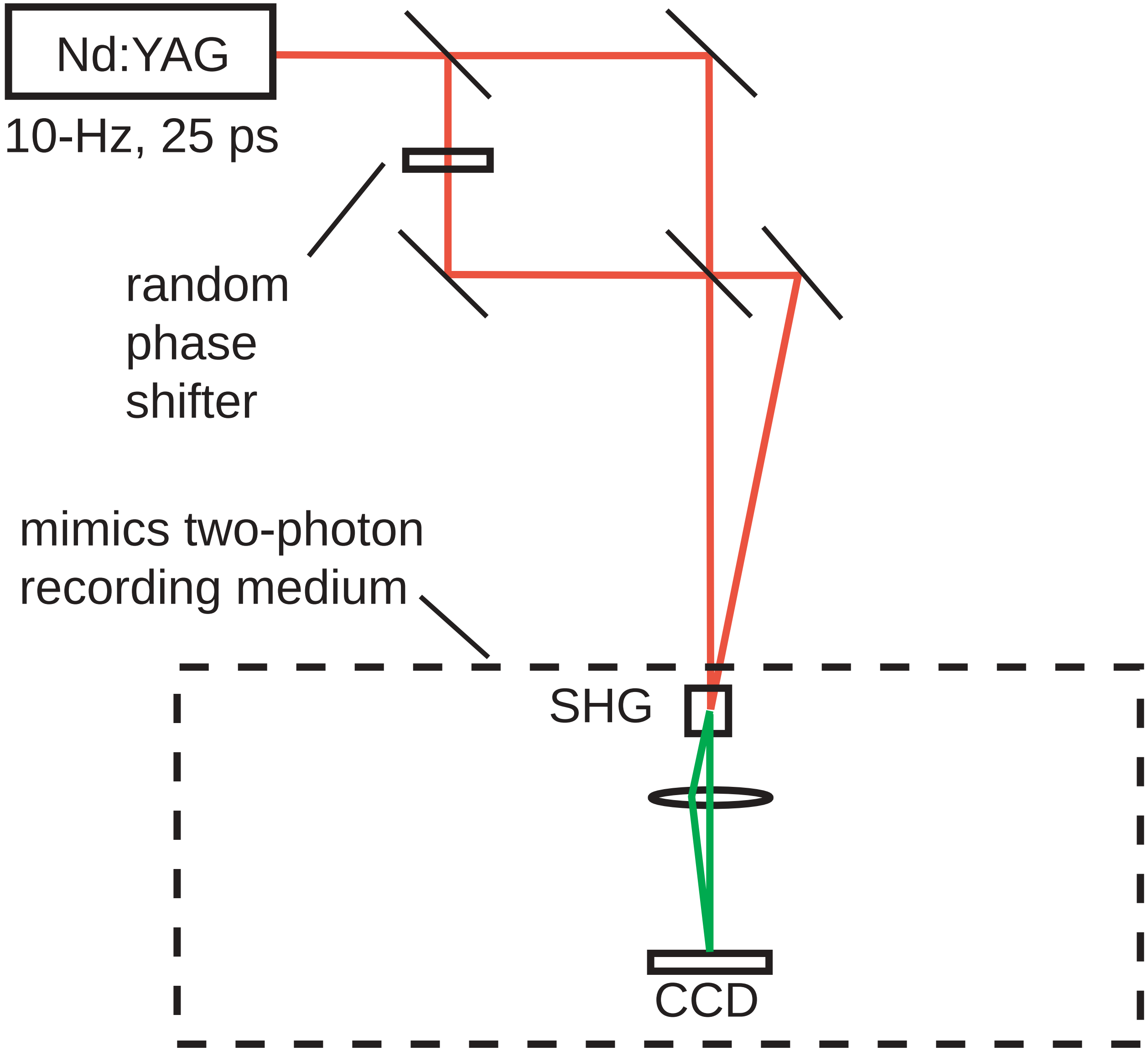
QUANTUM LITHOGRAPHY RESEARCH

Experimental Layout

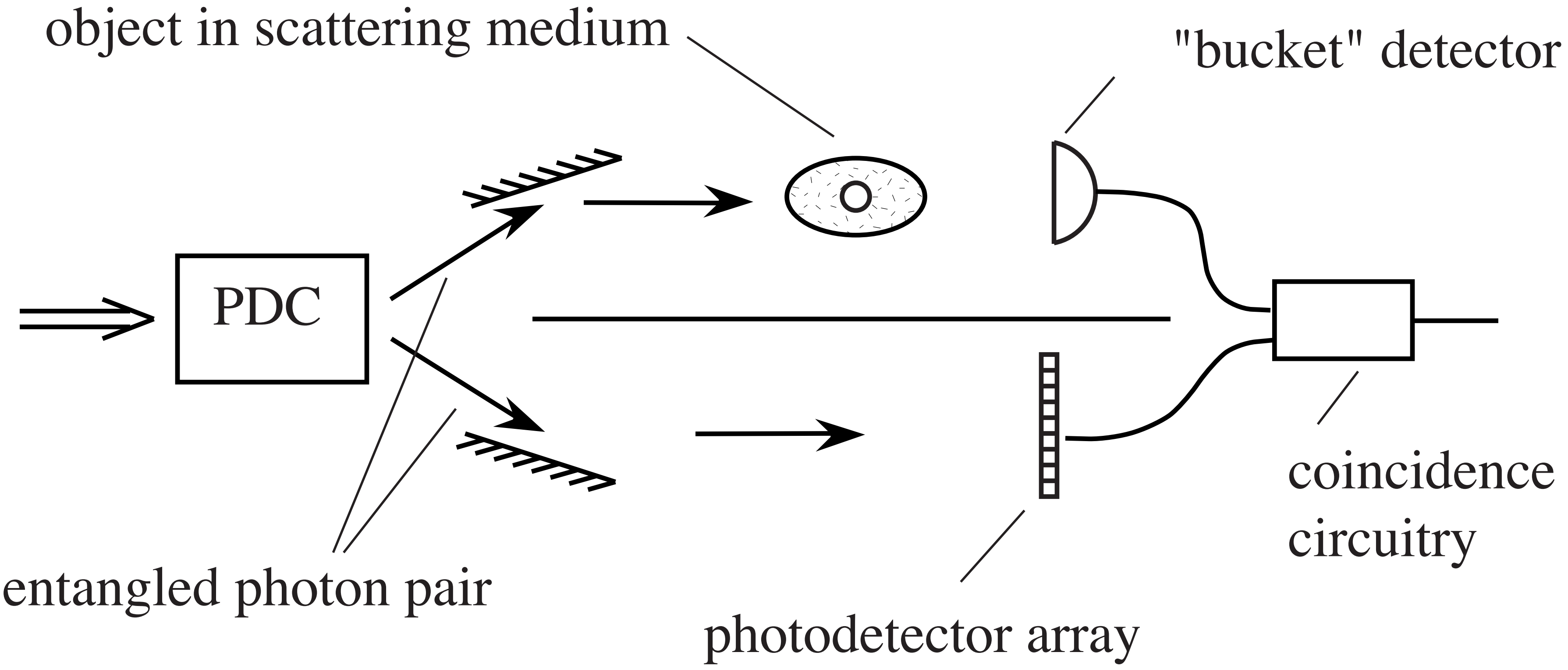




Classical Sub-Rayleigh Lithography Setup



Quantum (?) Coincidence Imaging

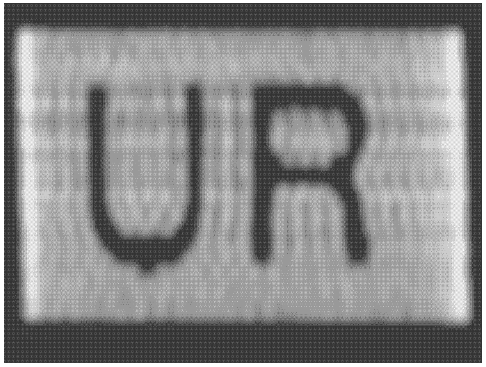


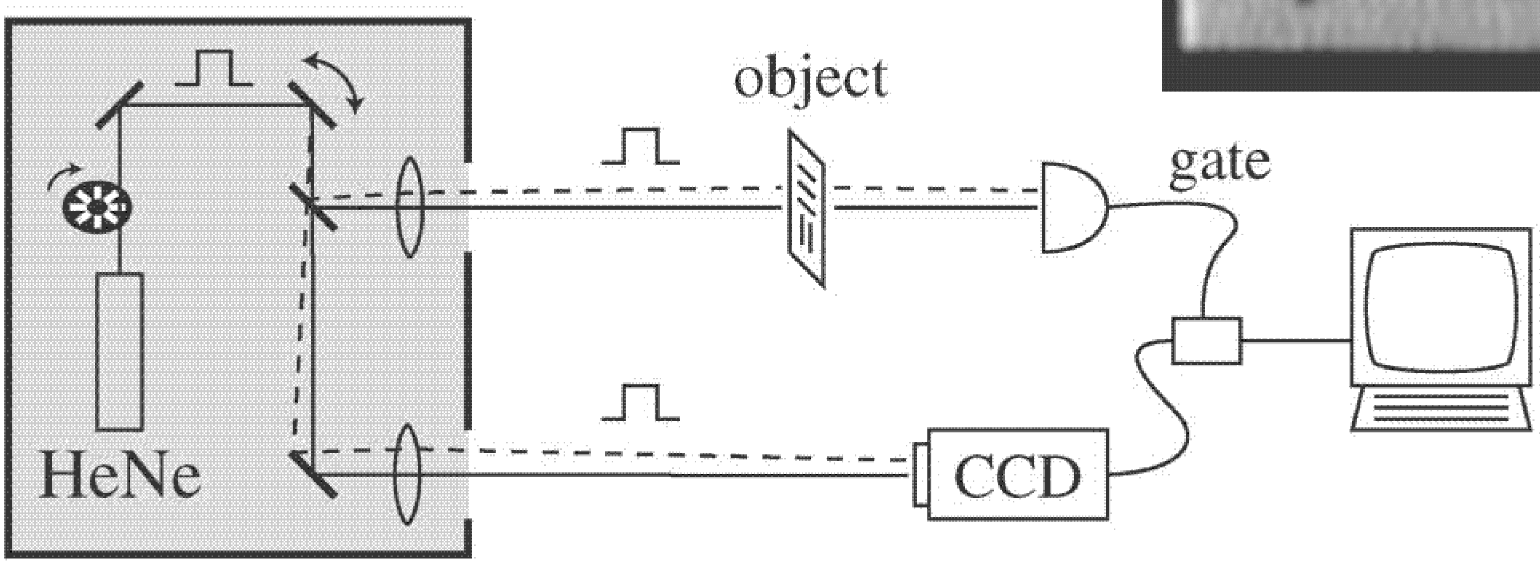
Obvious applicability to remote sensing!

Strekalov et al., Phys. Rev. Lett. **74**, 3600 (1995).
Pittman et al., Phys. Rev. A **52** R3429 (1995).
Abouraddy et al., Phys. Rev. Lett. **87**, 123602 (2001).

Classical Coincidence Imaging

We have performed coincidence imaging with a demonstrably classical source.

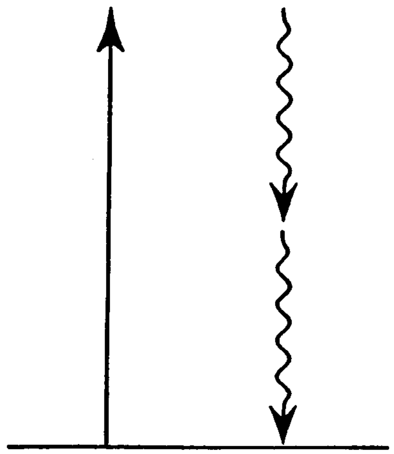




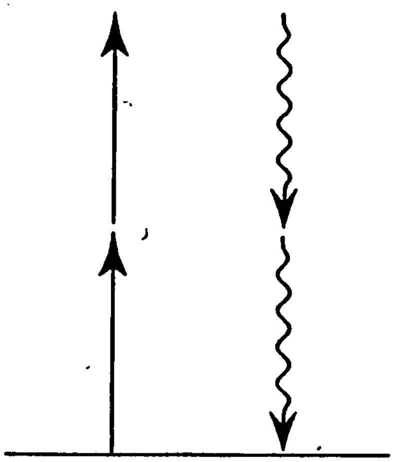
Bennink, Bentley, and Boyd, Phys. Rev. Lett. **89** 113601(2002).

TWO ROUTES TO ENTANGLEMENT

$\chi^{(2)}$



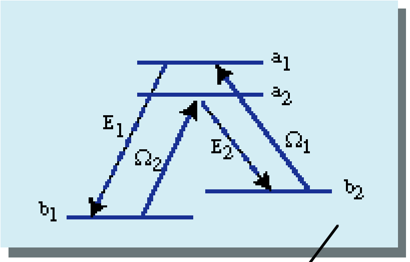
$\chi^{(3)}$



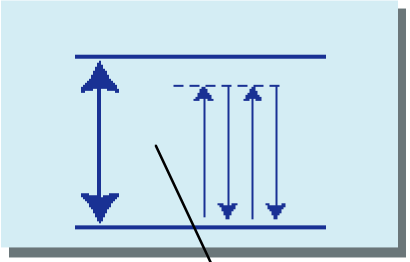
Generation of Quantum States of Light by Use of Electromagetically Induced Transparency

Robert W. Boyd and C. R. Stroud, Jr., University of Rochester

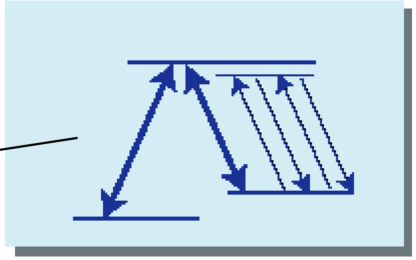
- Quantum states of light useful for applications including precision measurements and secure communications
- EIT enables the efficient creation of quantum states of light by eliminating spontaneous emission background noise.



double lambda EIT



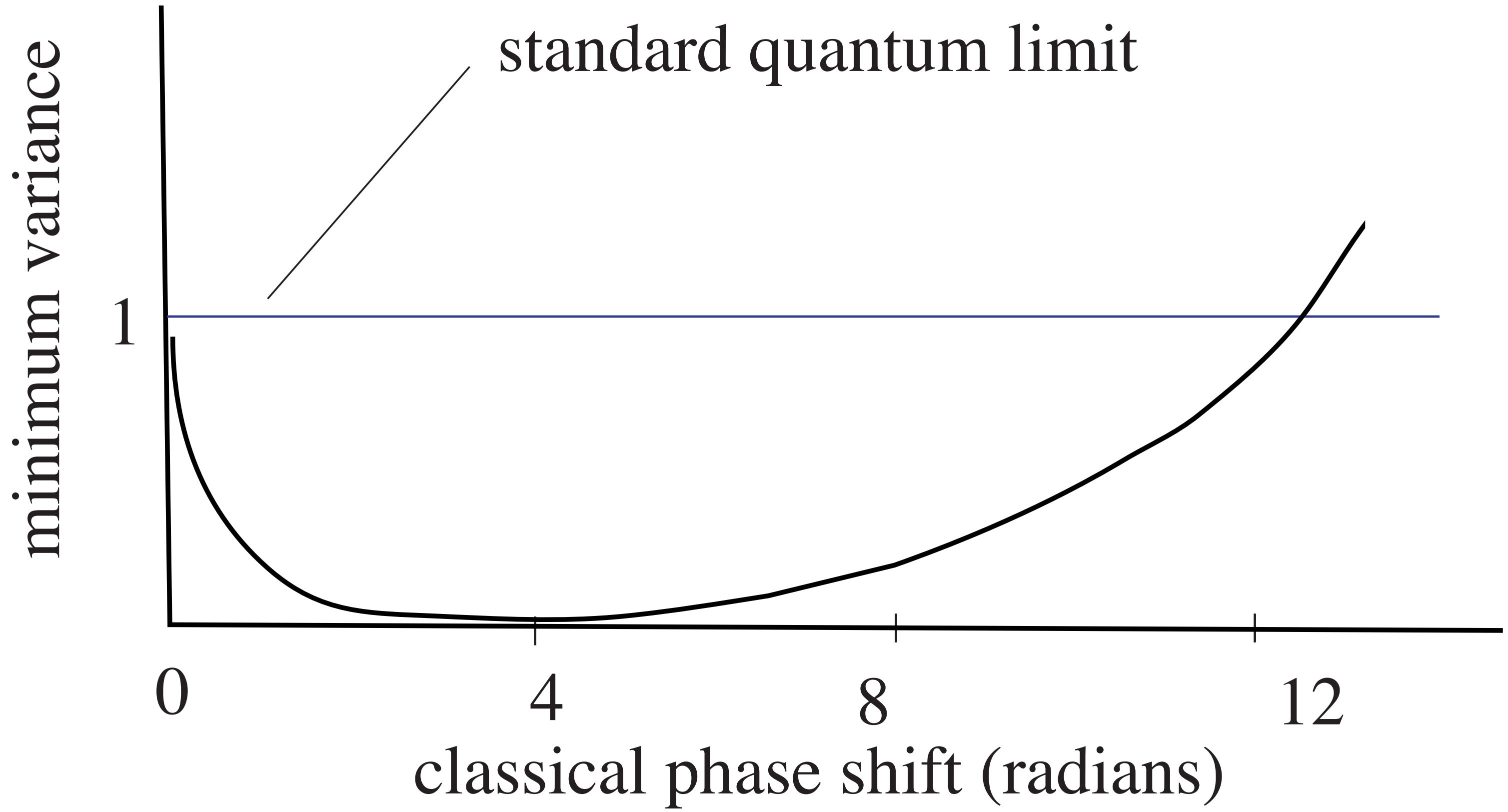
two-level EIT



dark-state EIT

Application of EIT to Squeezed-Light Generation

- Squeezing by self-phase modulation

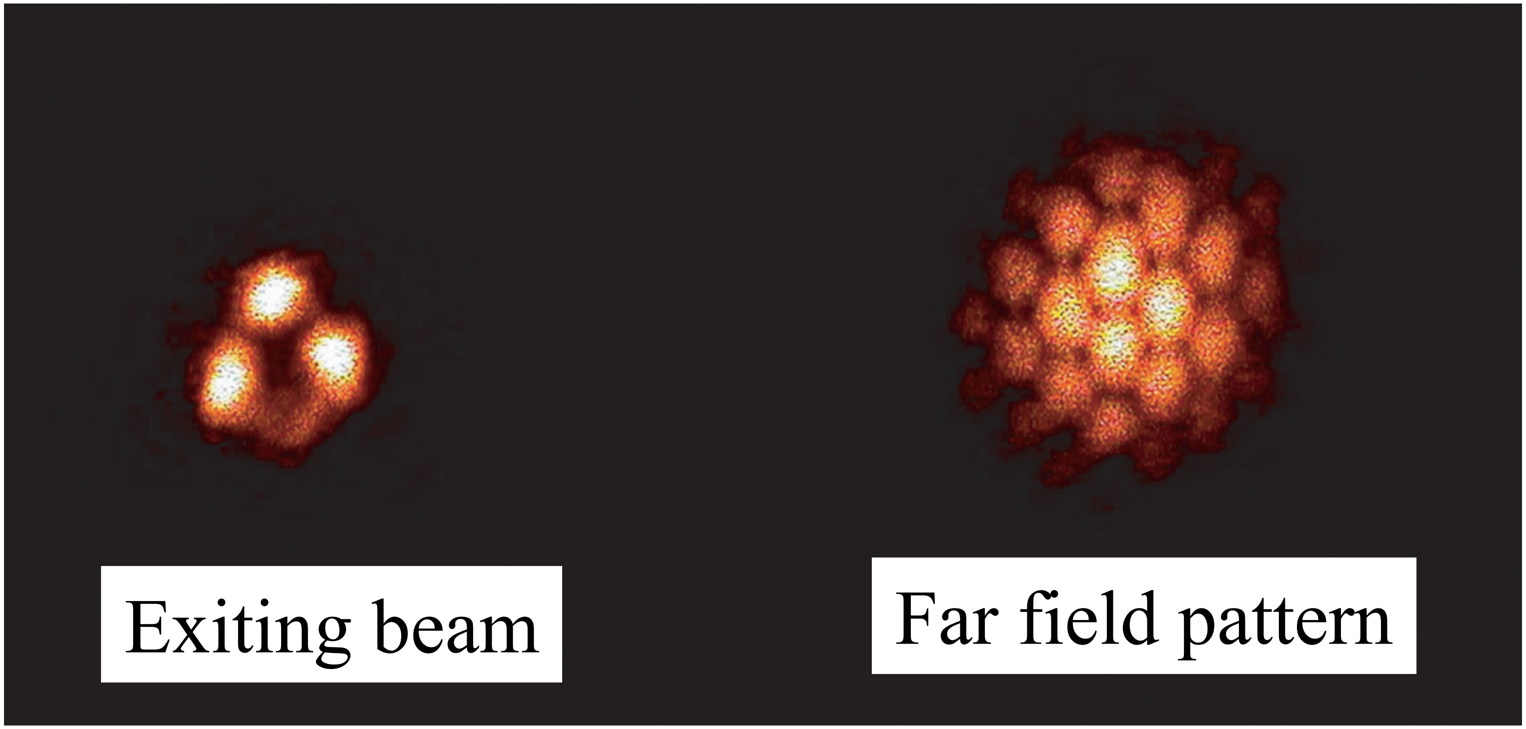


Blow, Loudon, and Phoenix, J. Mod. Opt., 40, 2515, 1993

- EIT allows phase shifts large enough to produce significant squeezing, and prevents signal-beam absorption which can degrade the squeezing.

Honey Comb Pattern Formation

Output from cell with single gaussian beam input



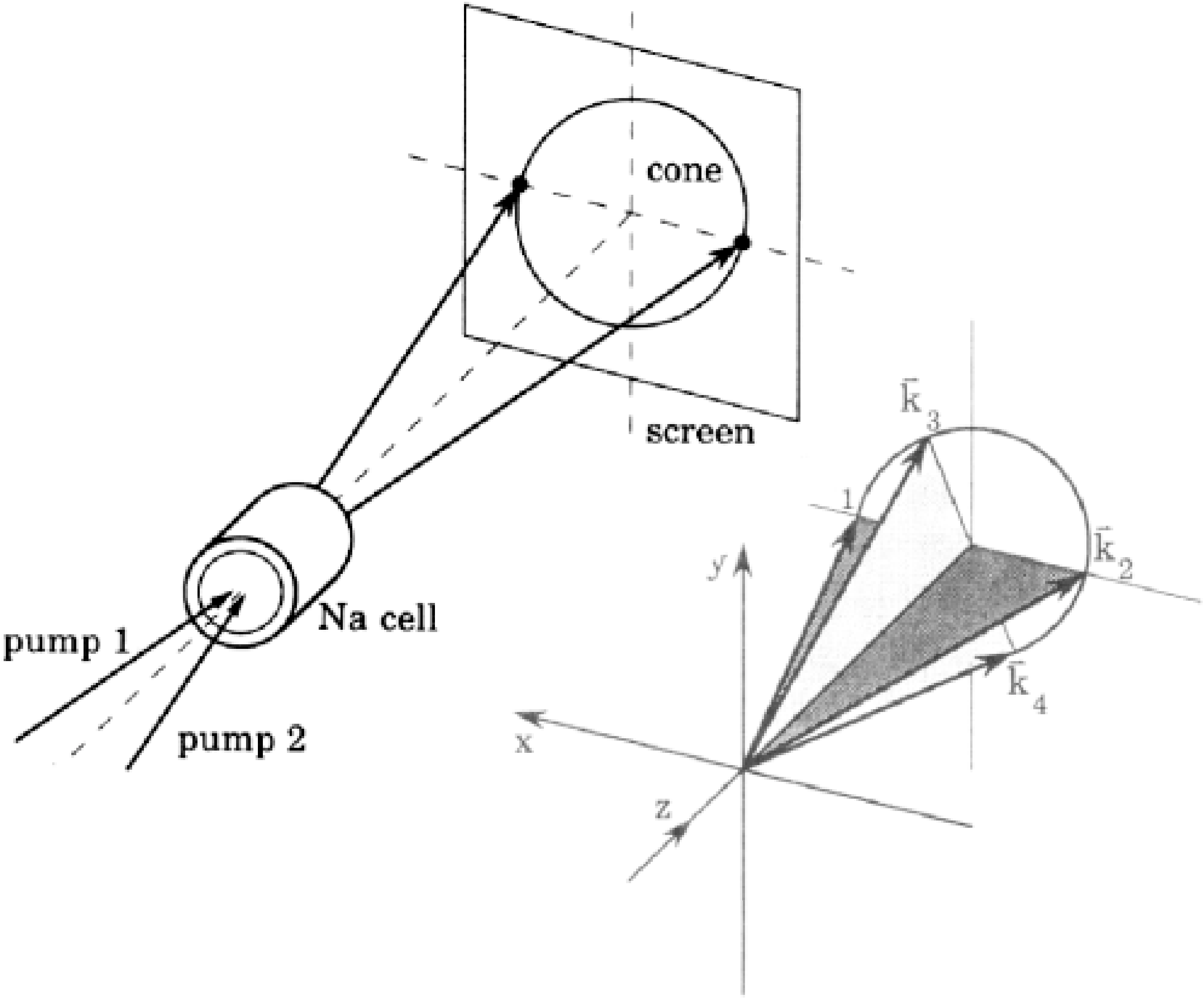
Quantum image?

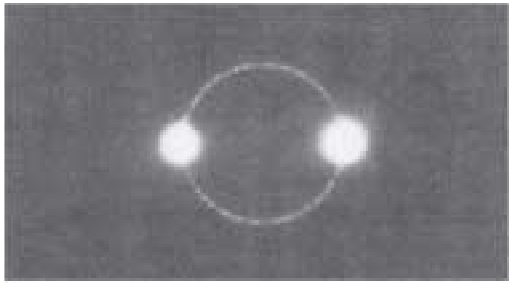
Input power 150 mW
Input beam diameter 0.22 mm
$\lambda = 588.995$ nm

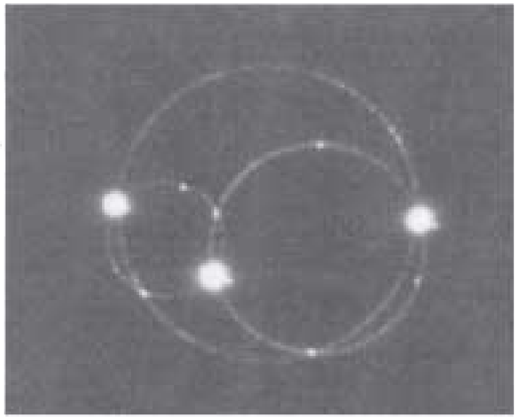
Sodium vapor cell T = 220° C

Bennink et al., PRL 88, 113901 2002.

Generation of Quantum States of Light by Two-Beam Excited Conical Emission

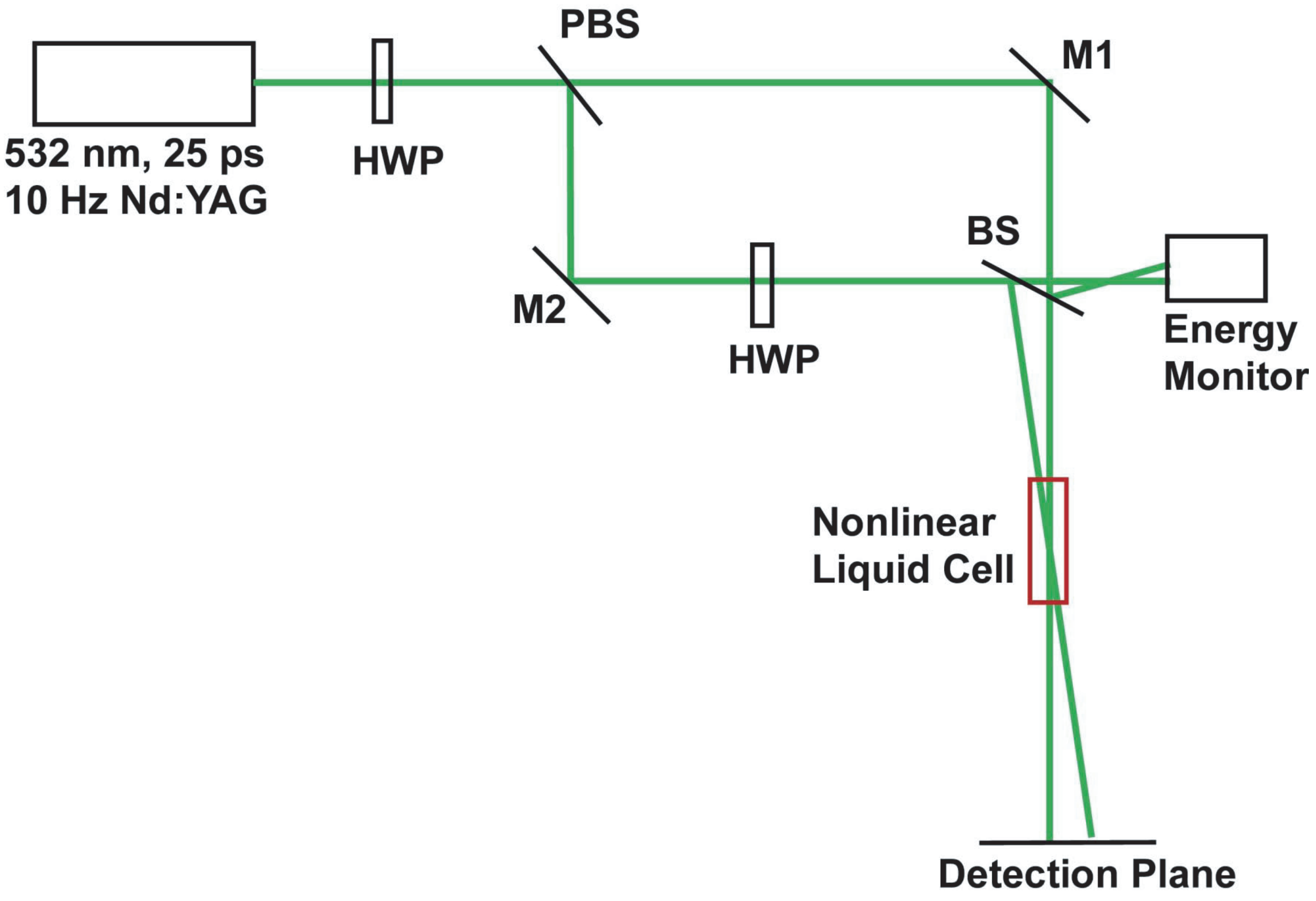






Kauranen et al, Opt. Lett. 16, 943, 1991; Kauranen and Boyd, Phys. Rev. A, 47, 4297, 1993.

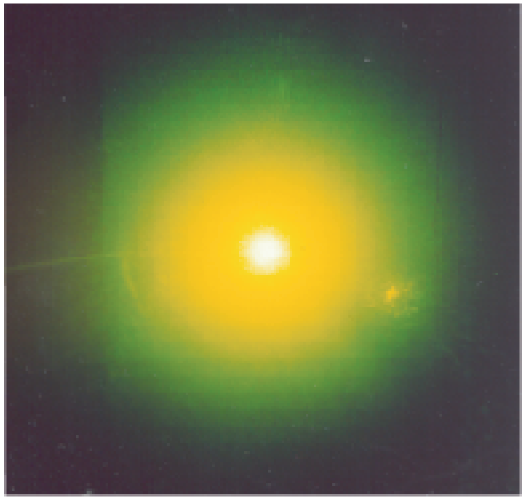
Experimental Configuration



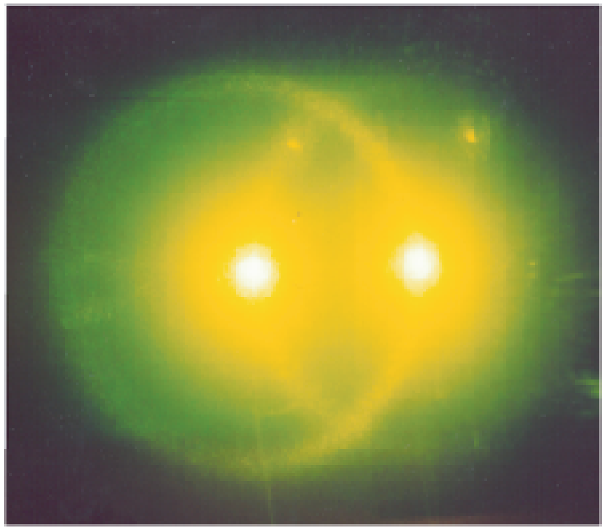
- Used 3-cm and 10-cm cells
- Used CS_2, CCl_4, and toluene
- Pulse intensities ~ 1-80 MW/cm^2
- Crossing angles ~ 0.003-0.04 rad

Conical Emission Patterns

Single input beam

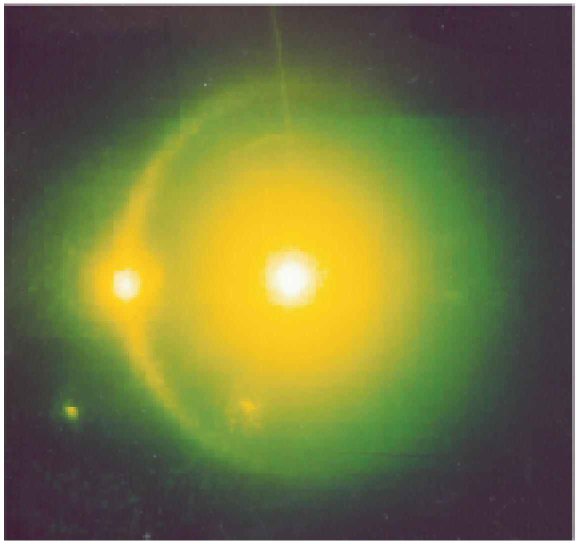


Two input beams
(equal intensity)
(parallel polarization)



Two cones formed, each centered on other beam.

Two input beams
(unequal intensity)
(parallel polarization)

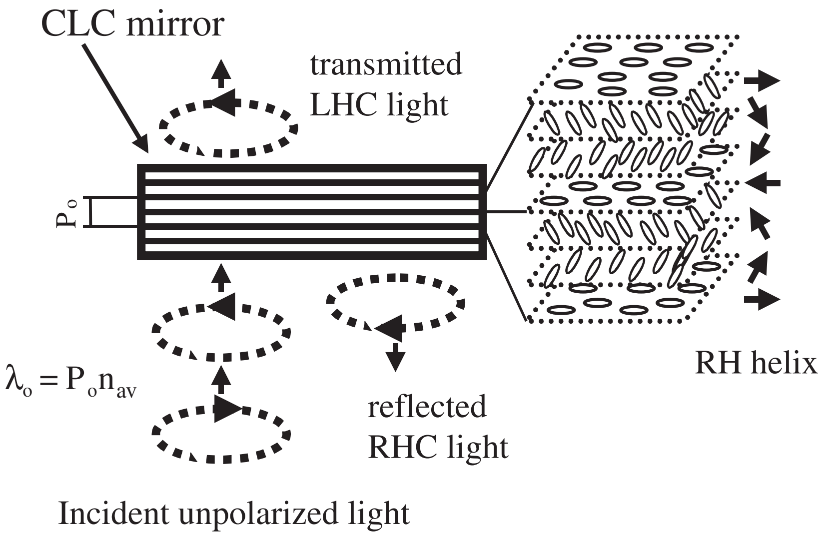


Only stronger input beam can act as pump for cone generation.

Generated in carbon disulfide

Source of Polarized, Single-Photons on Demand

- Useful for secure communication by quantum cryptgraphy
- Embed isolated dye molecules in chiral nematic liquid crystal
- Host acts as self-assembled photonic bandgap material
- Host composition helps prevent dye from bleaching
- Fluorescence shows strong antibunching


CLC mirror

transmitted
LHC light

P_o

RH helix

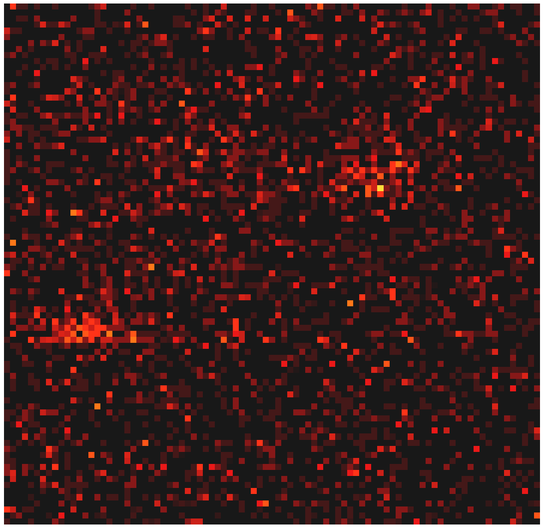
$\lambda_o = P_o n_{av}$

reflected
RHC light

Incident unpolarized light

Experimental procedure

Implementation with S. Lukishova


Single-molecule fluorescence

The Promise of Nonlinear Optics

Nonlinear optical techniques hold great promise for applications including:

- Photonic Devices
- Quantum Imaging
- Quantum Computing/Communications
- Optical Switching
- Optical Power Limiters
- All-Optical Image Processing

But the lack of high-quality photonic materials is often the chief limitation in implementing these ideas.

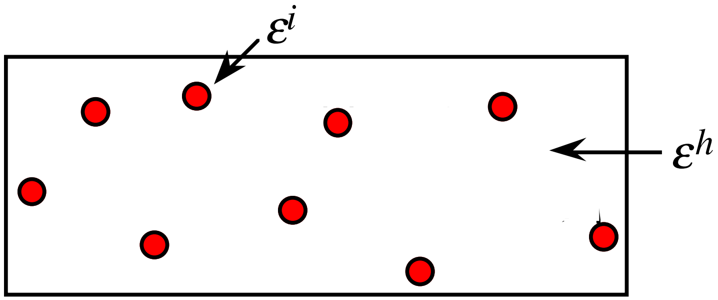
Approaches to the Development of Improved NLO Materials

- New chemical compounds
- Quantum coherence (EIT, etc.)
- Composite Materials:
  - (a) Microstructured Materials, e.g.
    - Photonic Bandgap Materials,
    - Quasi-Phasematched Materials, etc
  - (b) Nanocomposite Materials

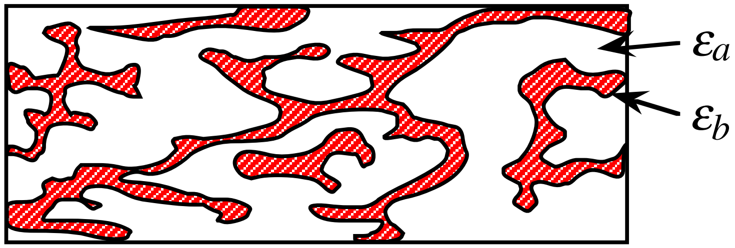
These approaches are not incompatible and in fact can be exploited synergistically!

Nanocomposite Materials for Nonlinear Optics

- Maxwell Garnett

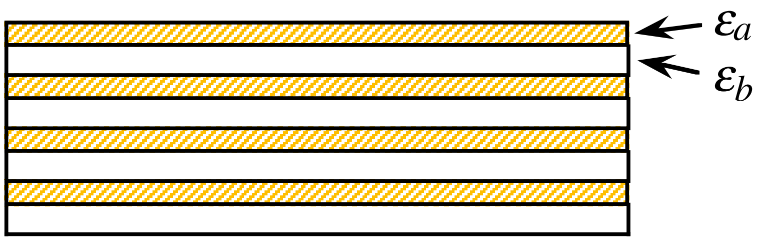


- Bruggeman (interdispersed)



- Fractal Structure

- Layered



scale size of inhomogeneity << optical wavelength

Gold-Doped Glass

A Maxwell-Garnett Composite

gold volume fraction approximately 10^{-6}

gold particles approximately 10 nm diameter

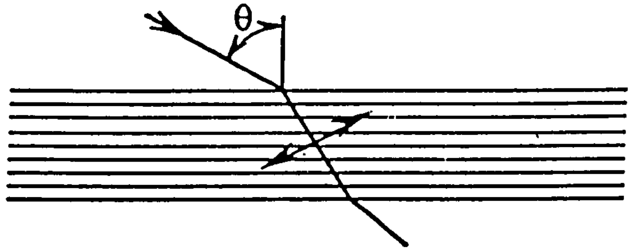
- Composite materials can possess properties very different from their constituents.

- Red color is because the material absorbs very strongly at the surface plasmon frequency (in the blue) -- a consequence of local field effects.

Demonstration of Enhanced NLO Response

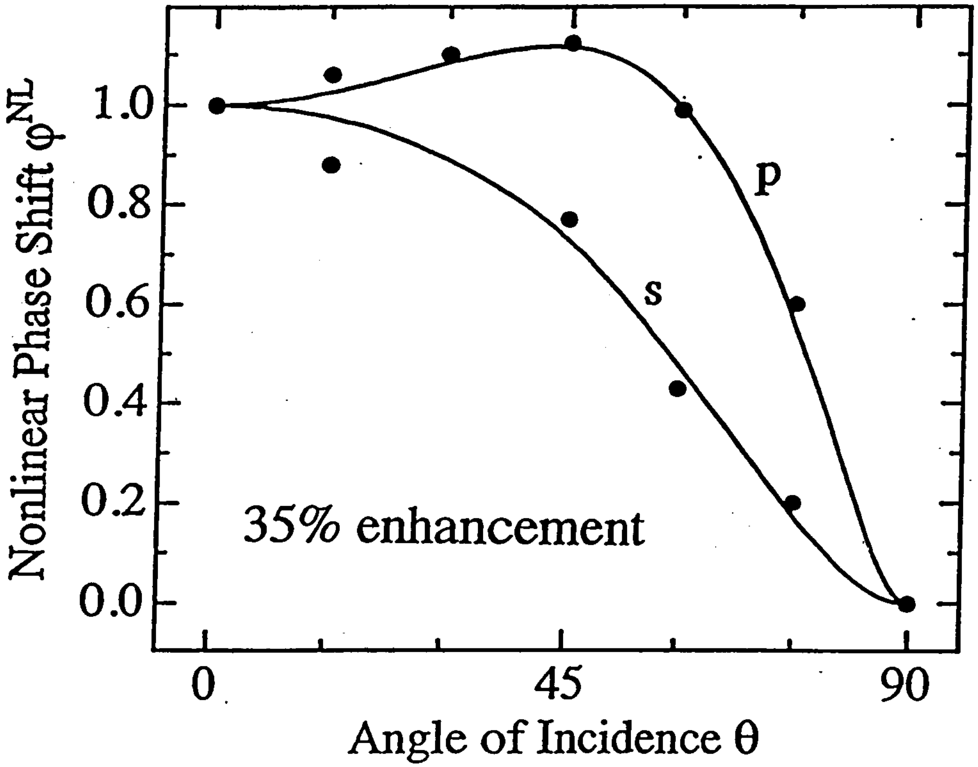
- Alternating layers of TiO_2 and the conjugated polymer PBZT.

- Measure NL phase shift as a function of angle of incidence



$\nabla \cdot \mathbf{D} = 0$ implies that $(\varepsilon \mathbf{E})_\perp$ is continuous.

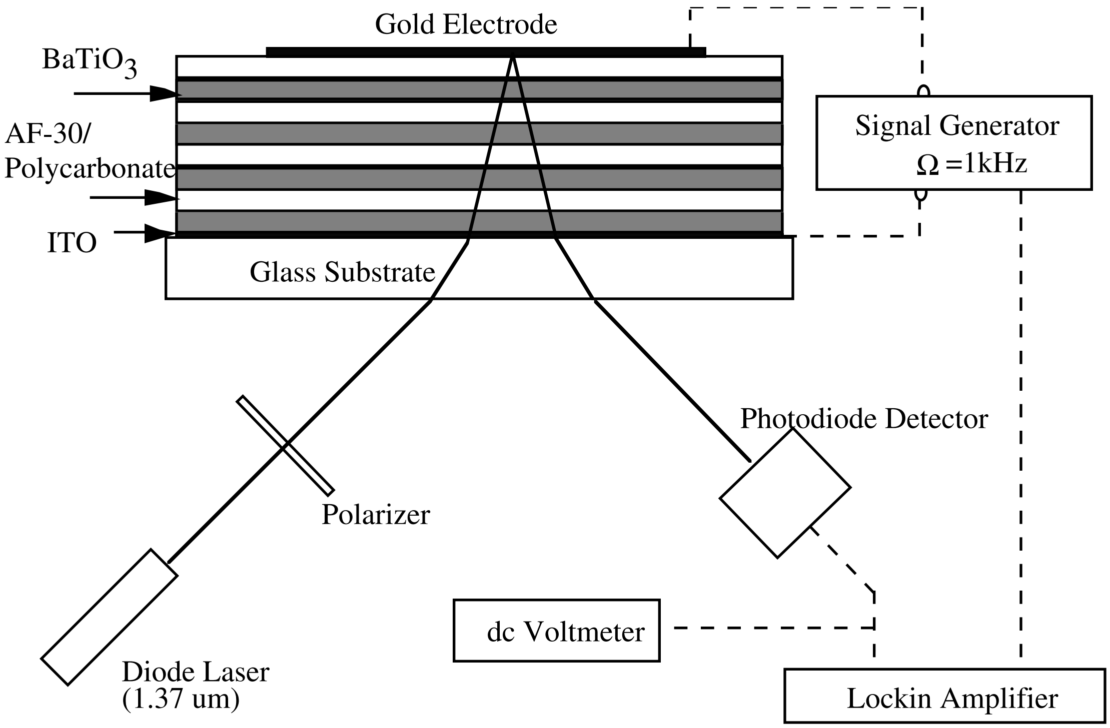
Thus field is concentrated in *lower* index material.



Fischer, Boyd, Gehr, Jenekhe, Osaheni, Sipe, and Weller-Brophy, Phys. Rev. Lett. 74, 1871, 1995.

Gehr, Fischer, Boyd, and Sipe, Phys. Rev. A 53, 2792 1996.

Enhanced EO Response of Layered Composite Materials



$$\chi_{ijkl}^{(eff)}(\omega';\omega,\Omega_1,\Omega_2) = f_a\left[\frac{\varepsilon_{eff}(\omega')}{\varepsilon_a(\omega')}\right]\left[\frac{\varepsilon_{eff}(\omega)}{\varepsilon_a(\omega)}\right]\left[\frac{\varepsilon_{eff}(\Omega_1)}{\varepsilon_a(\Omega_1)}\right]\left[\frac{\varepsilon_{eff}(\Omega_2)}{\varepsilon_a(\Omega_2)}\right]\chi_{ijkl}^{(a)}(\omega';\omega,\Omega_1,\Omega_2)$$

- AF-30 (10%) in polycarbonate (spin coated)
  n=1.58 ε(dc) = 2.9
- barium titante (rf sputtered)
  n=1.98 ε(dc) = 15

$$\chi_{zzzz}^{(3)} = (3.2 + 0.2i) \times 10^{-21}\,(m/V)^2 \pm 25\%$$

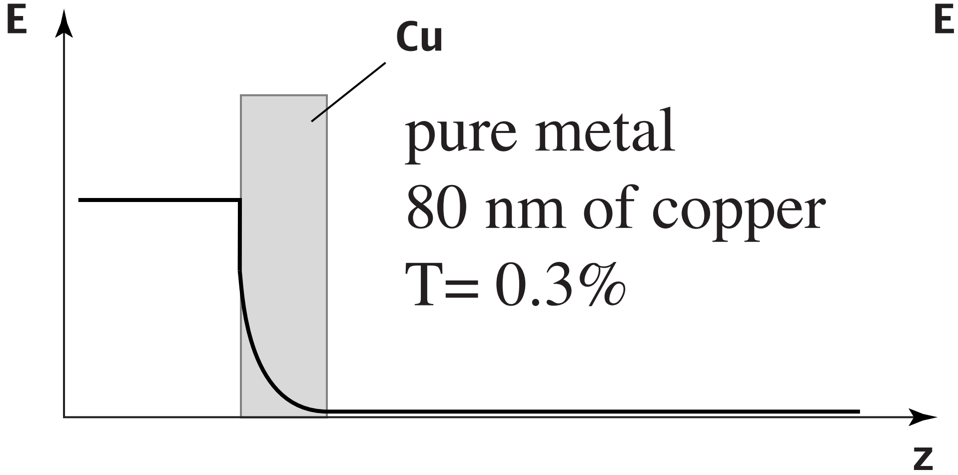
$$\approx 3.2\,\chi_{zzzz}^{(3)}(\text{AF-30 / polycarbonate})$$

3.2 times enhancement in agreement with theory

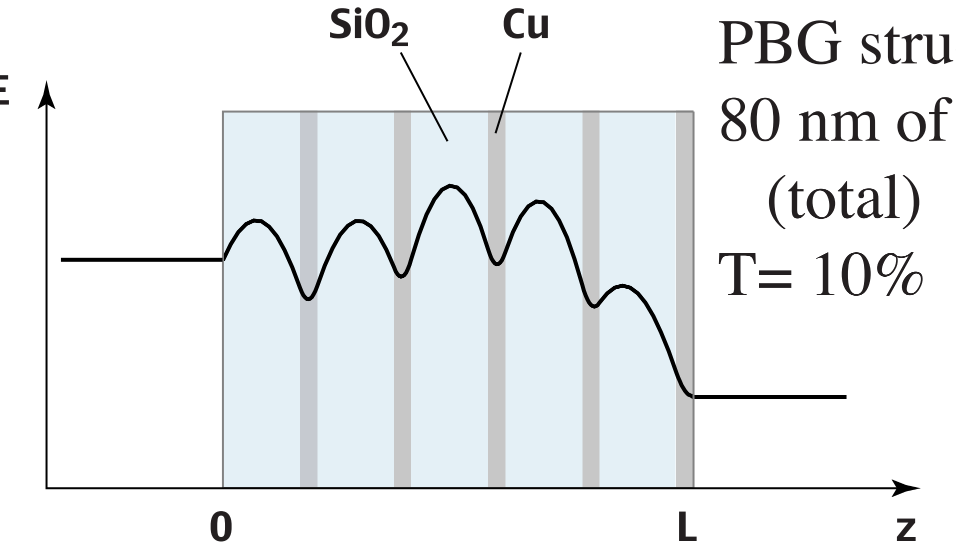
R. L. Nelson, R. W. Boyd, Appl. Phys. Lett. 74, 2417, 1999.

Accessing the Optical Nonlinearity of Metals with Metal-Dielectric PBG Structures

- Metals have very large optical nonlinearities but low transmission.
- Low transmission is because metals are highly reflecting (not because they are absorbing!).
- Solution: construct metal-dielectric PBG structure.
  (linear properties studied earlier by Bloemer and Scalora)



pure metal
80 nm of copper
T= 0.3%



PBG structure
80 nm of Cu
(total)
T= 10%

40 times enhancment of NLO response is predicted!

R.S. Bennink, Y.K. Yoon, R.W. Boyd, and J. E. Sipe Opt. Lett. 24, 1416, 1999.

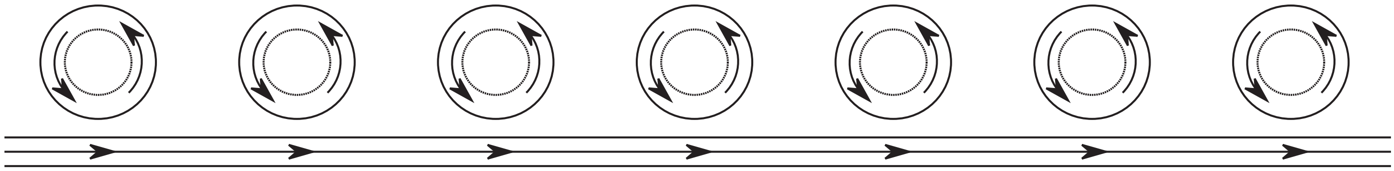
Nanofabrication

- Materials (artificial materials)
- Devices

(distinction?)

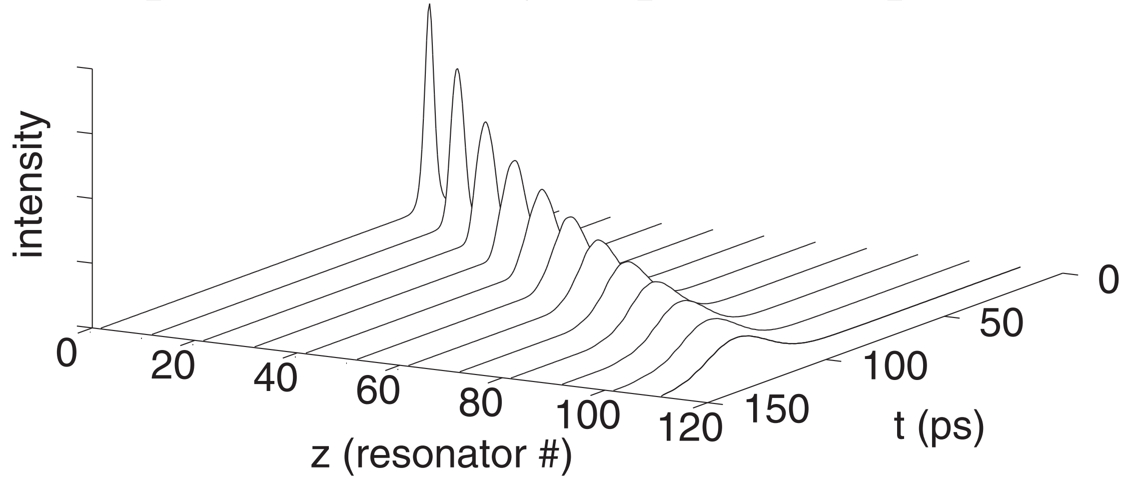
NLO of SCISSOR Devices

(Side-Coupled Integrated Spaced Sequence of Resonators)

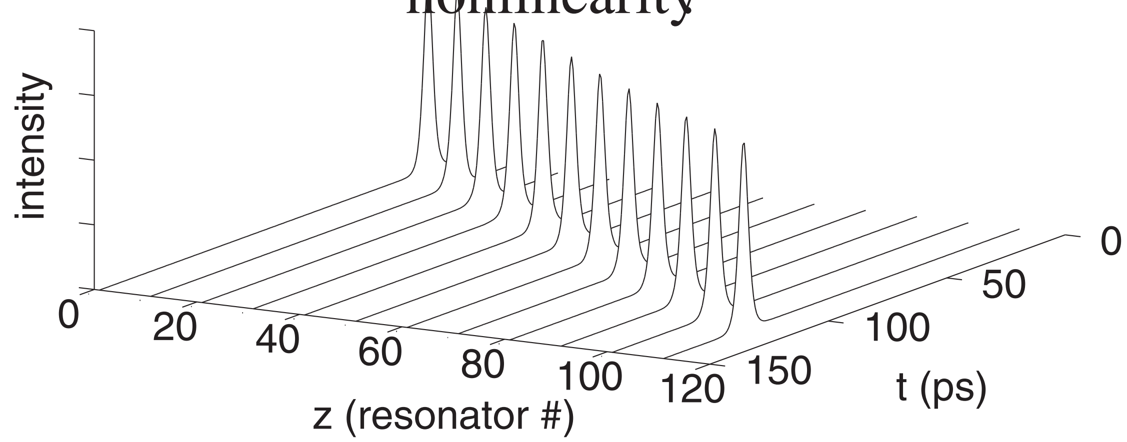


Displays slow-light, tailored dispersion, and optical solitons.
Description by NL Schrodinger eqn. in continuum limit.

- Pulses spread when only dispersion is present



- But form solitons through balance of dispersion and nonlinearity



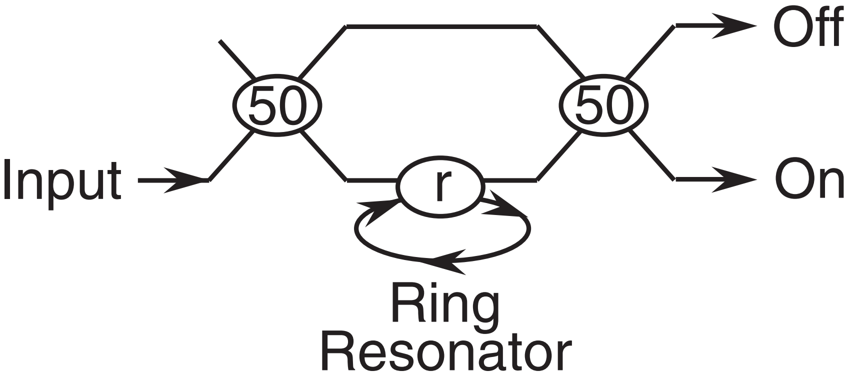
(J.E. Heebner, Q-Han Park and RWB)

Ultrafast All-Optical Switch Based On Arsenic Triselenide Chalcogenide Glass

- We excite a whispering gallery mode of a chalcogenide glass disk.

- The nonlinear phase shift scales as the square of the finesse F of the resonator. (F ≈ 10^2 in our design)

- Goal is 1 pJ switching energy at 1 Tb/sec.



J. E. Heebner and R. W. Boyd, Opt. Lett. 24, 847, 1999.
(implementation with Dick Slusher, Lucent)

Photonic Devices for Biosensing

Objective:

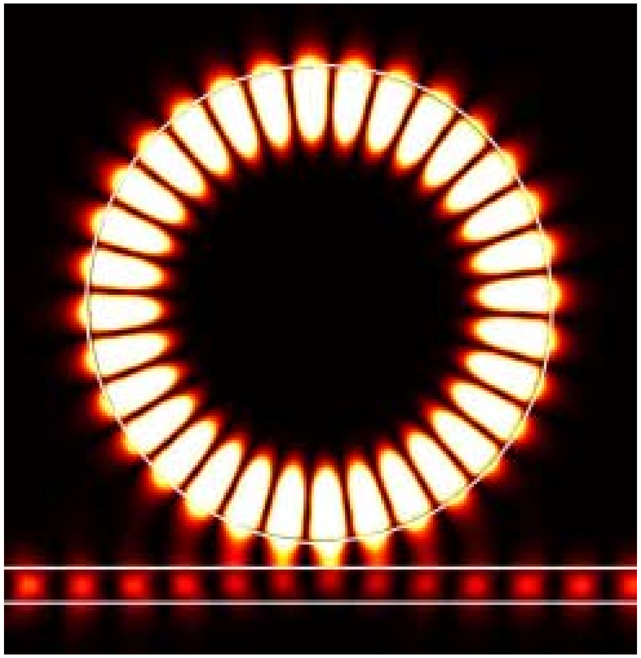
Obtain high sensitivity, high specificity detection of pathogens through optical resonance

Approach:

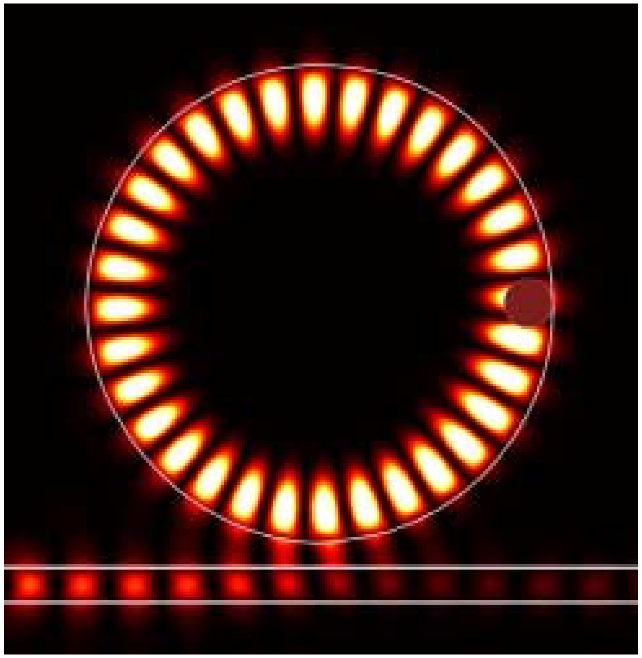
Utilize high-finesse whispering-gallery-mode disk resonator.

Presence of pathogen on surface leads to dramatic decrease in finesse.

Simulation of device operation:



Intensity distribution in absense of absorber.



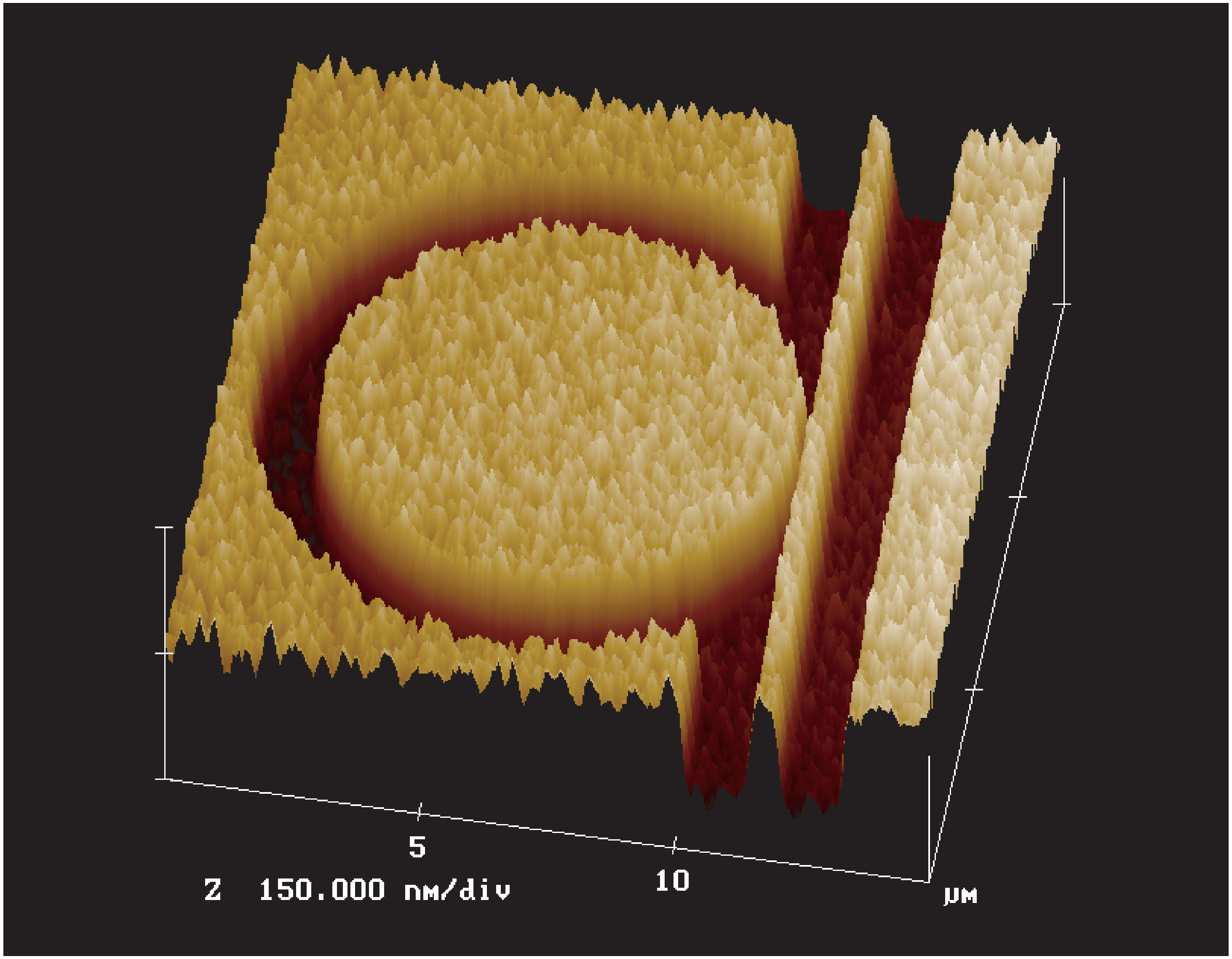
Intensity distribution in presence of absorber.

FDTD

A Real Whispering Gallery

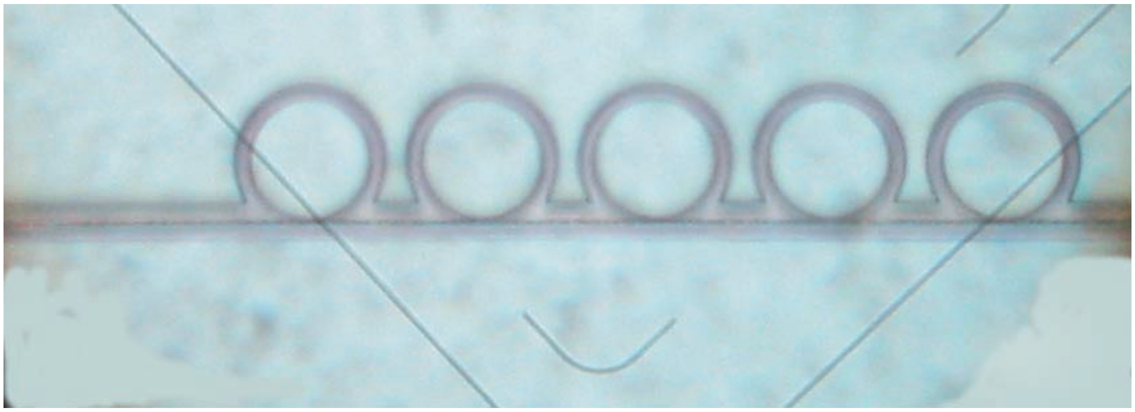
St. Paul's Cathedral, London

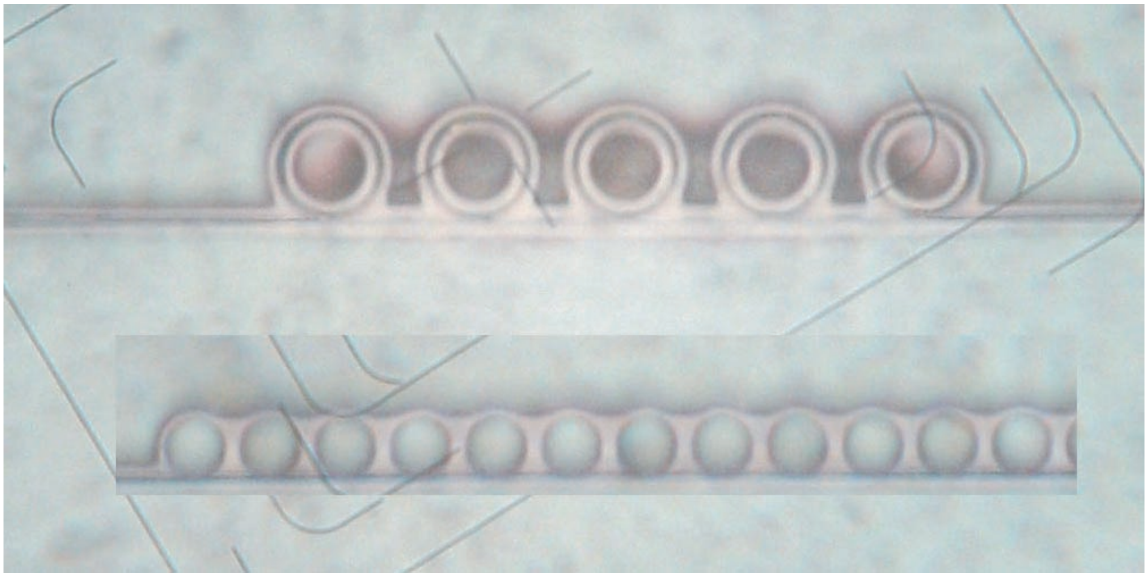
Disk Resonator and Optical Waveguide in PMMA Resist

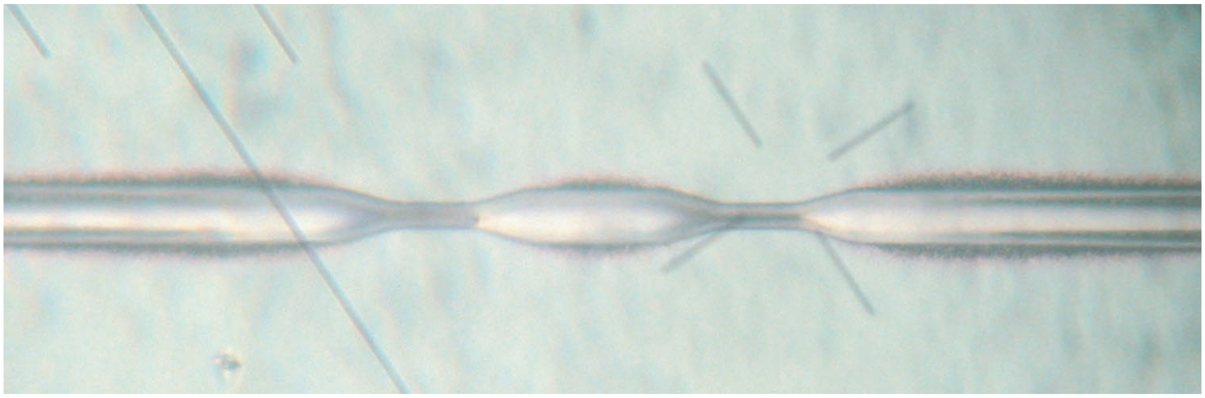


AFM

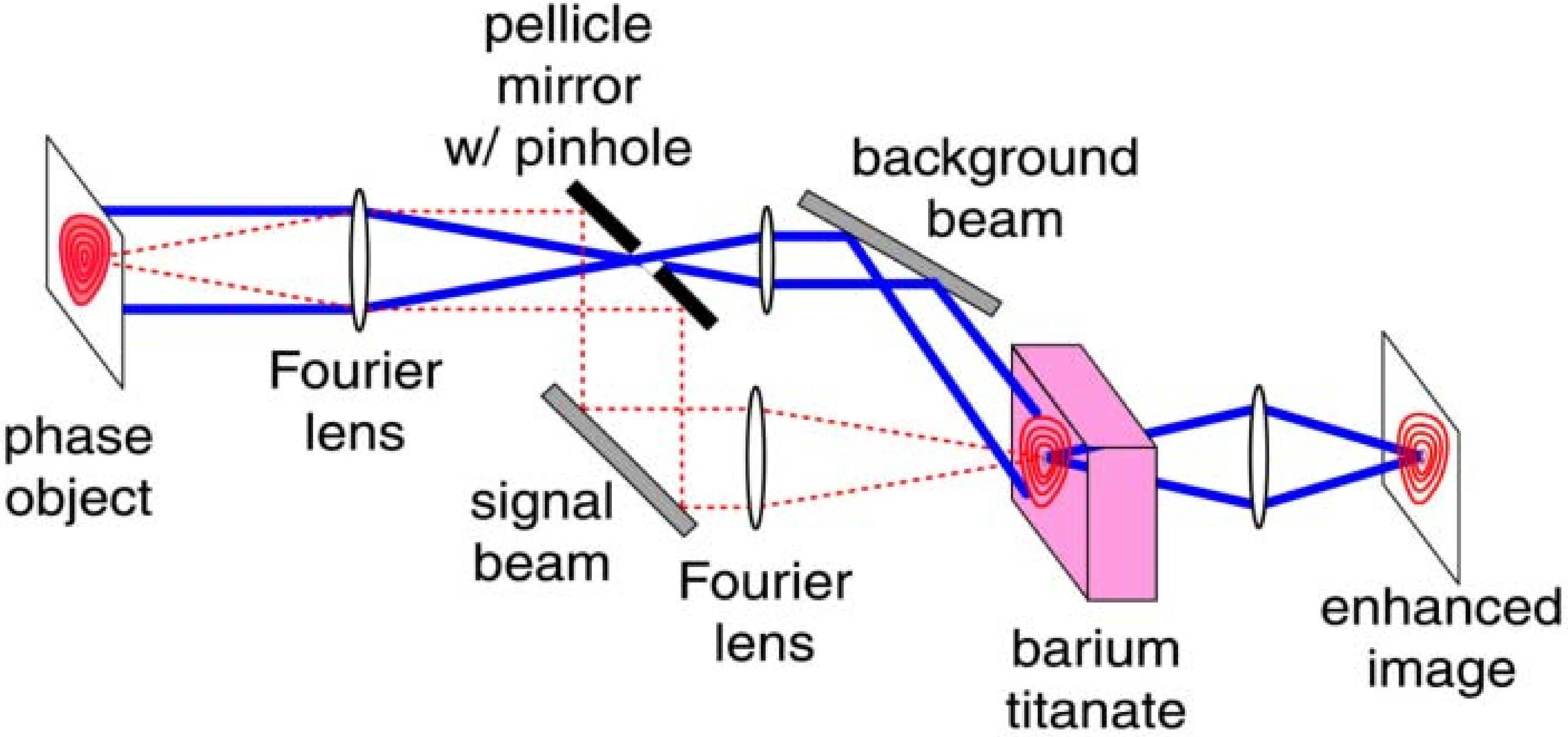
Photonic Devices in GaAs/AlGaAs





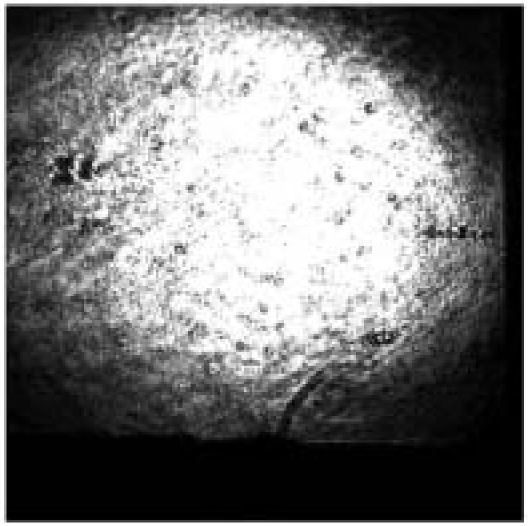


Nonlinear Optical Microscopy

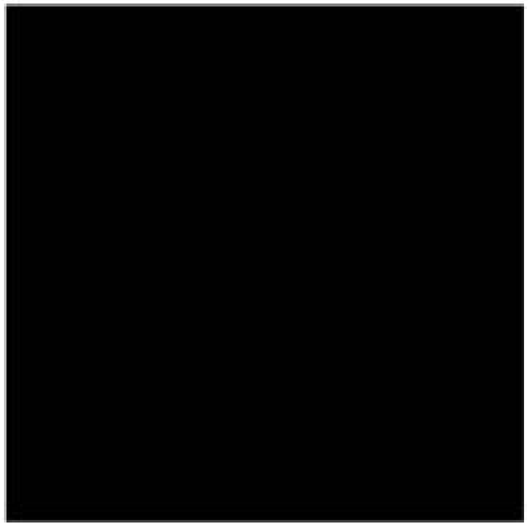


J. E. Heebner and R. W. Boyd, Optics Communications, 182, 243-247, 2000.

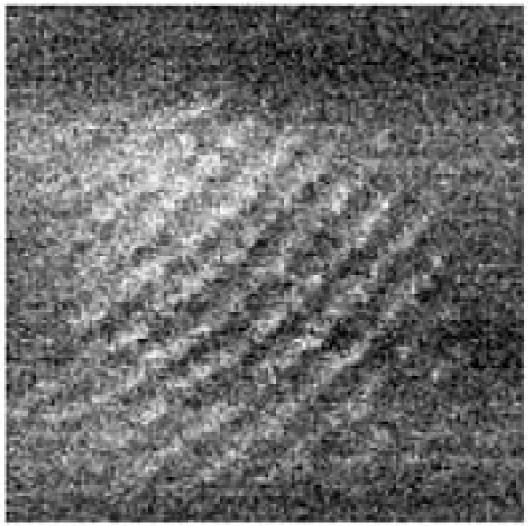
Fingerprint Enhancement



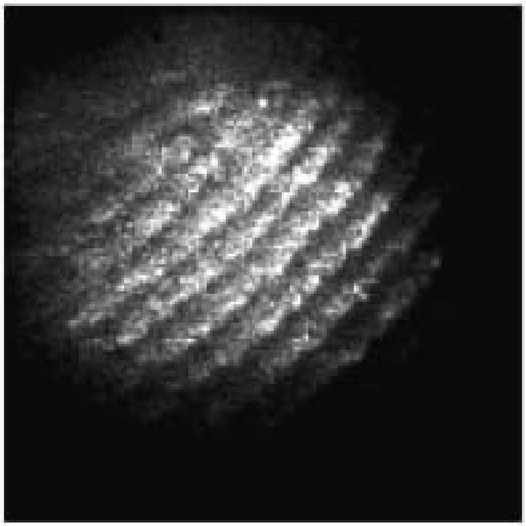
raw
image
(invisible)



filtered
image
(too weak!)

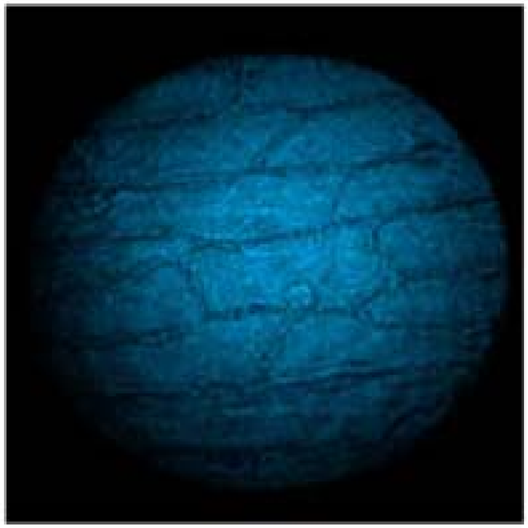


digitally
amplified
(noisy)



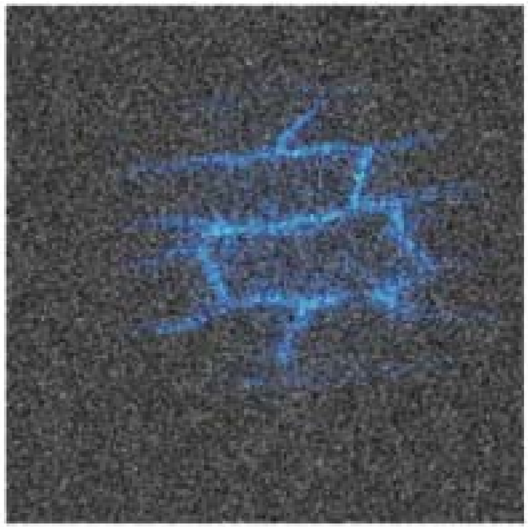
optically
pre-amplified
(PhORCE)

Onion Skin Cell Visualization

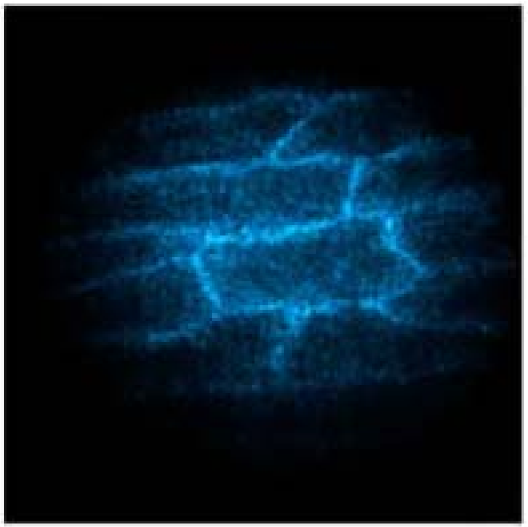


raw
image
(barely
visible)

filtered
image
(too weak!)

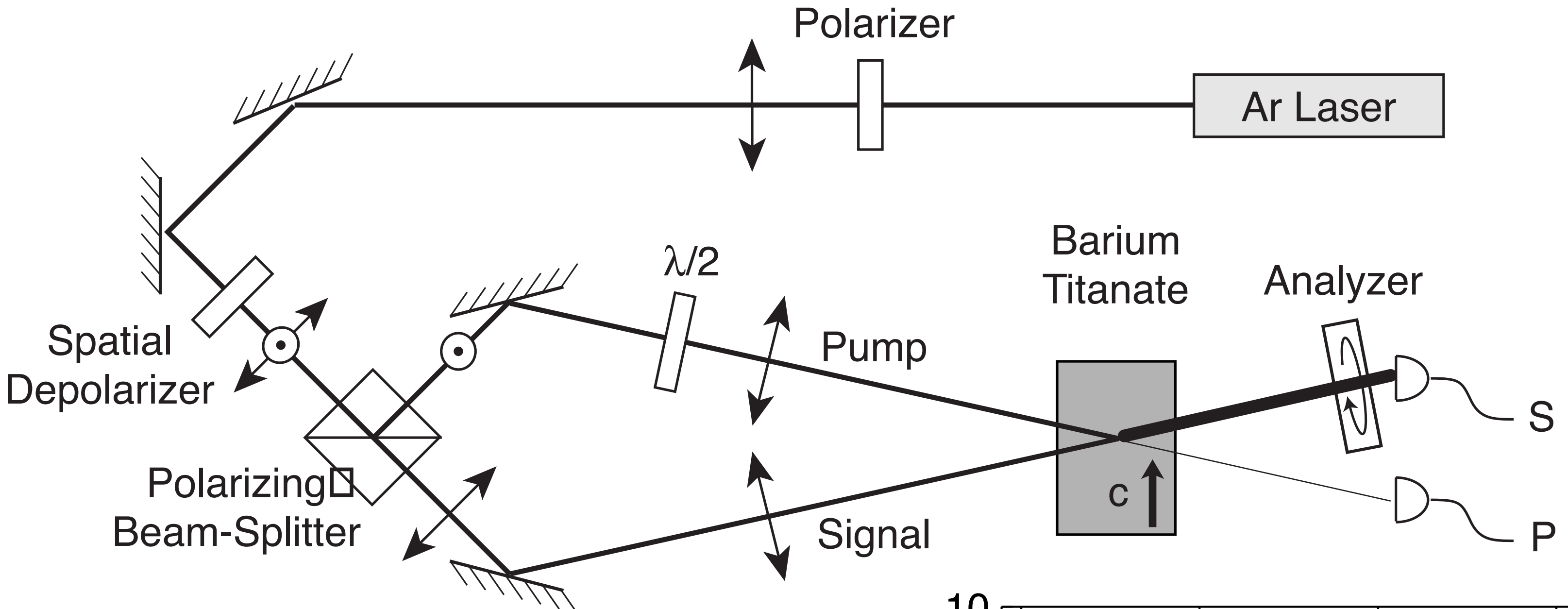


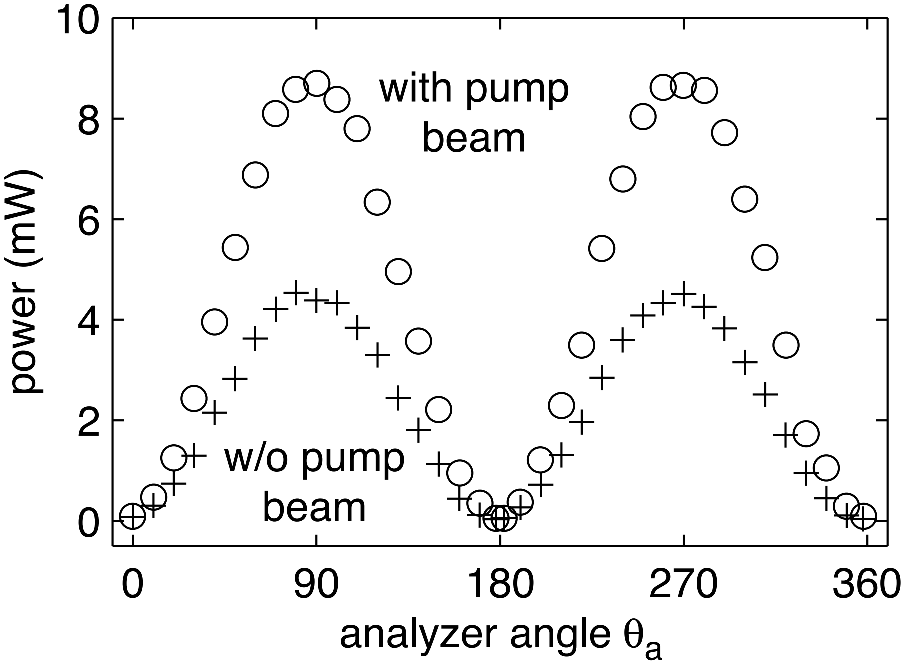
digitally
amplified
(noisy)



optically
pre-amplified
(PhORCE)

Construction of a Photorefractive Polarizer With Greater Than 50% Transmission



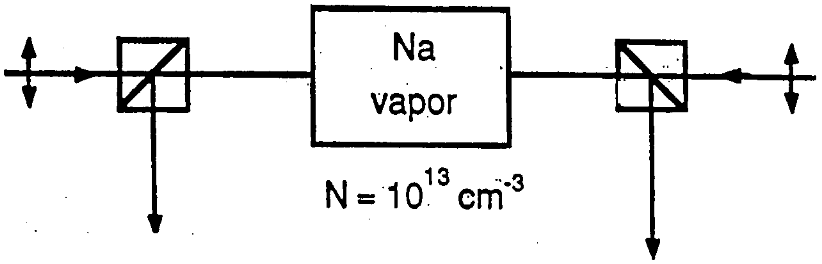


Heebner, Bennink, Boyd, and Fisher, Opt. Lett. 25, 257, 2000.

Some Underlying Issues in Nonlinear Optics

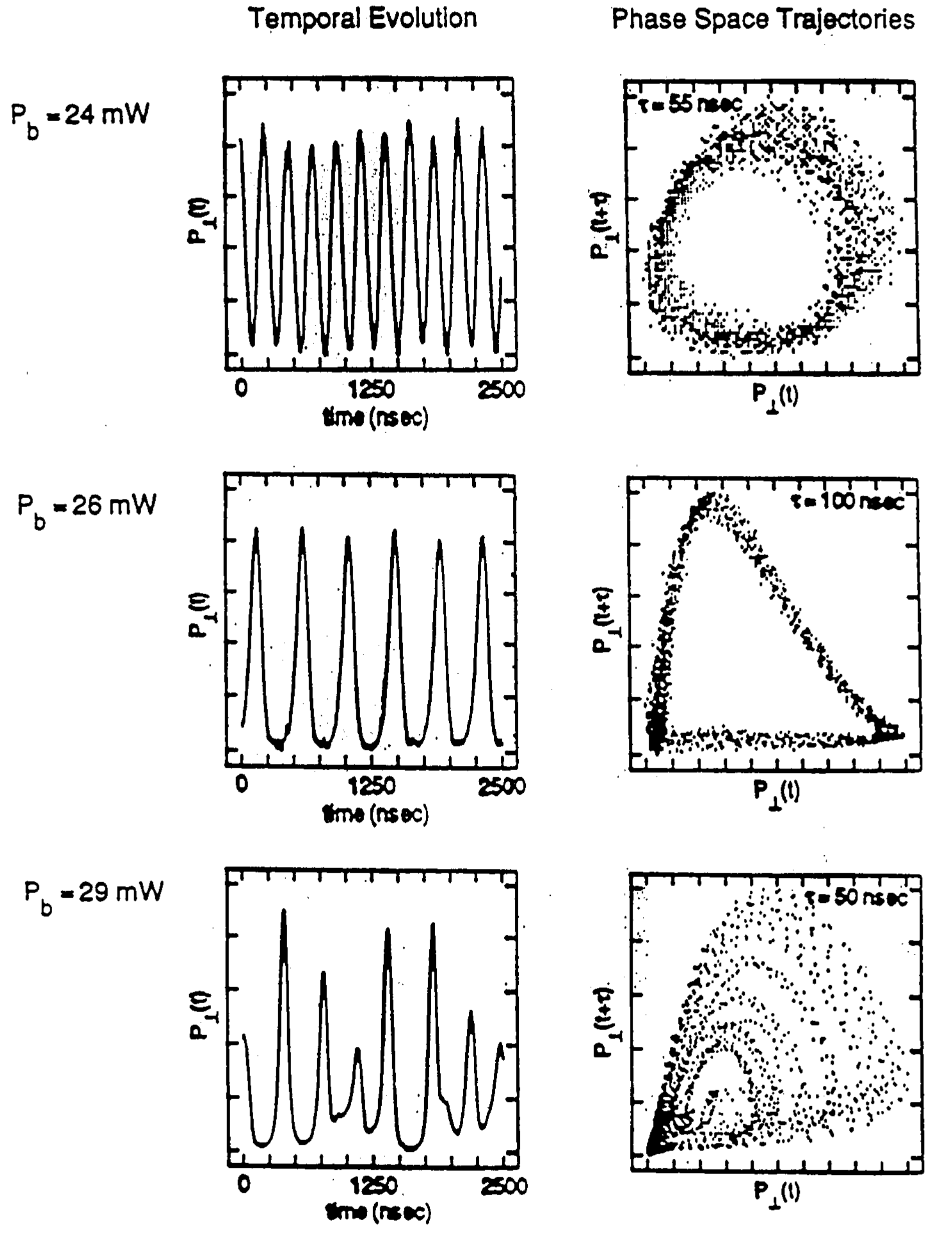
- Self-Assembly/Self-Organization in Nonlinear Systems
- Stability vs. Instability (and Chaos) in Nonlinear Systems

Chaos in Sodium Vapor



Na vapor

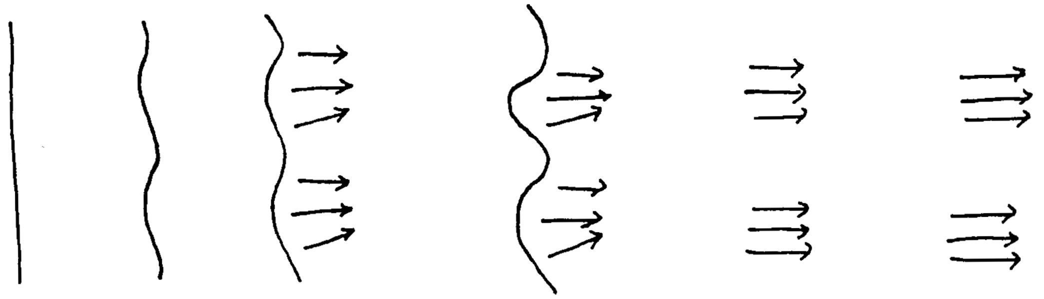
$N = 10^{13}\ cm^{-3}$



PRL 58, 2432 (1987); 61, 1827 (1988); 64 1721 (1990).

Laser Beam Filamentation

Spatial growth of wavefront perturbations



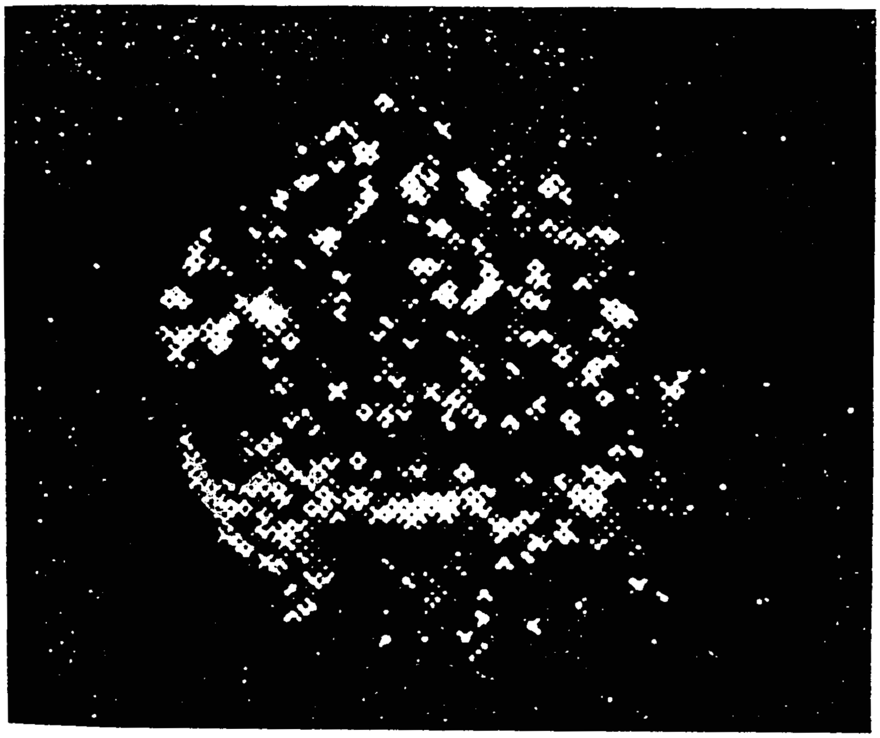
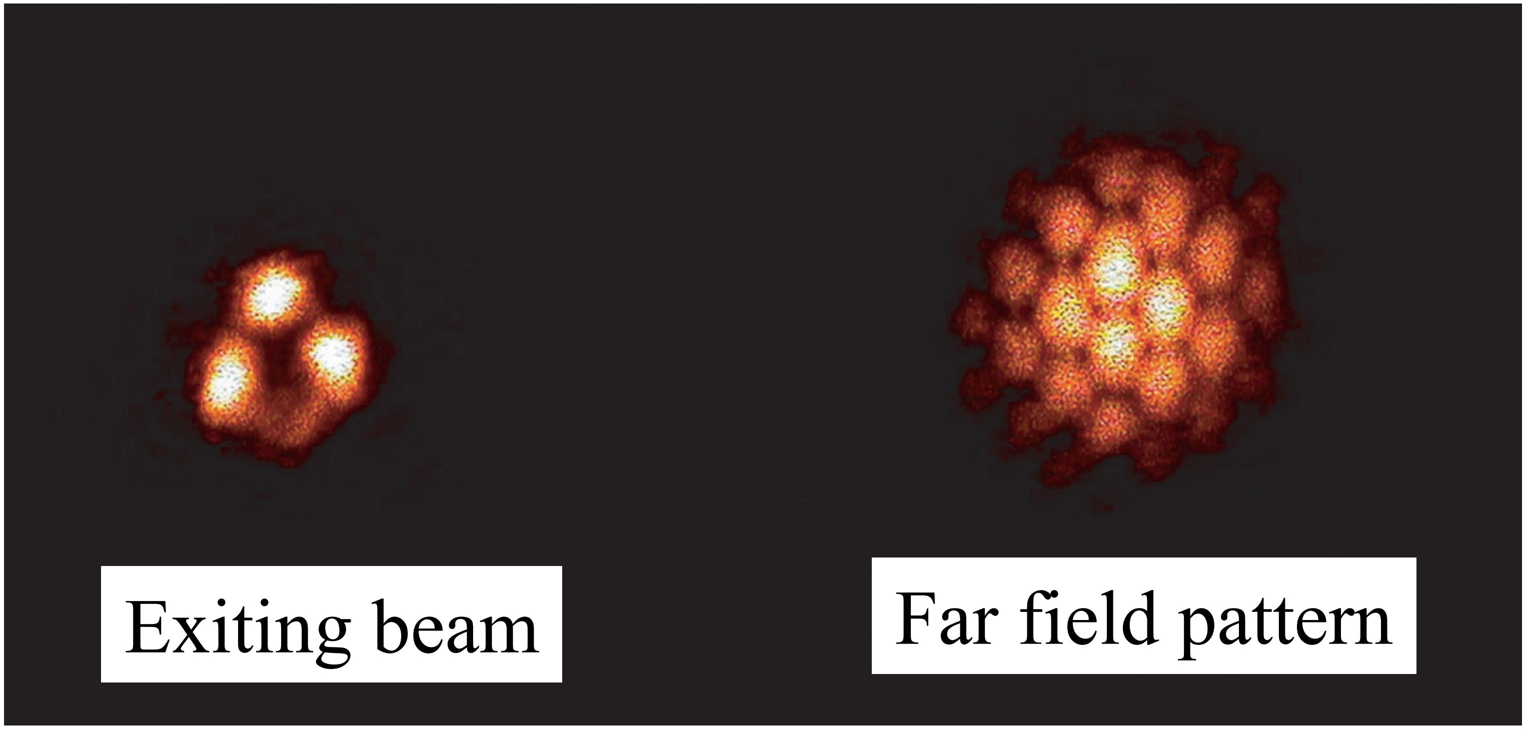


Fig. 17.2 Image of small-scale filaments at the exit windows of a CS_2 cell created by self-focusing of a multimode laser beam. [After S. C. Abbi and H. Mahr, *Phys. Rev. Lett.* 26, 604 (1971).]

Honey Comb Pattern Formation

Output from cell with single gaussian beam input



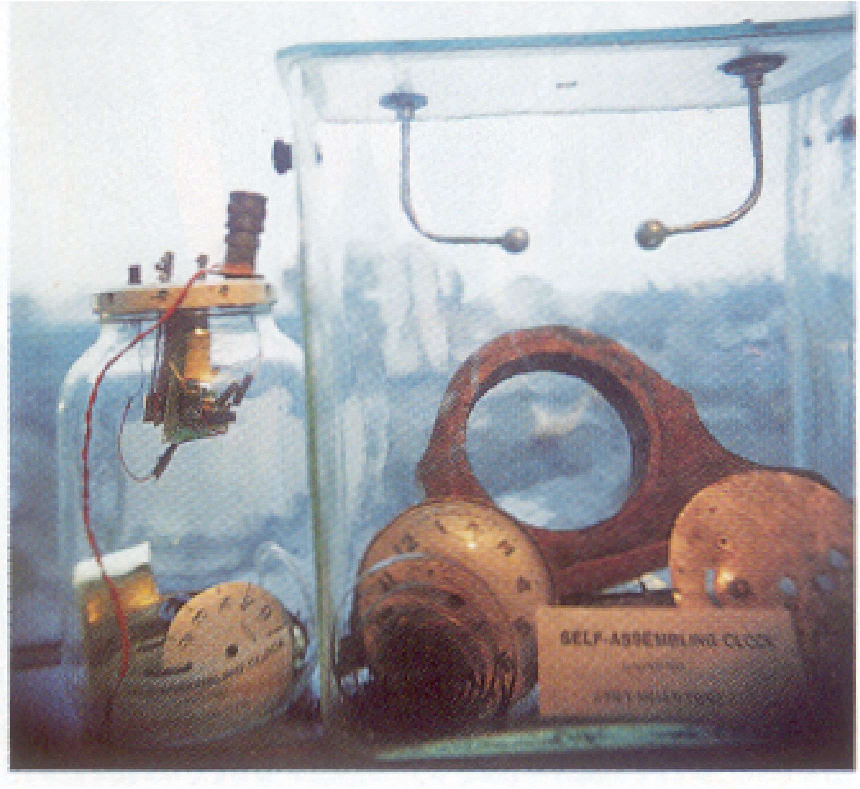
Quantum image?

Input power 150 mW
Input beam diameter 0.22 mm
$\lambda = 588.995$ nm

Sodium vapor cell T = 220° C

Bennink et al., PRL 88, 113901 2002.

Experiment in Self Assembly



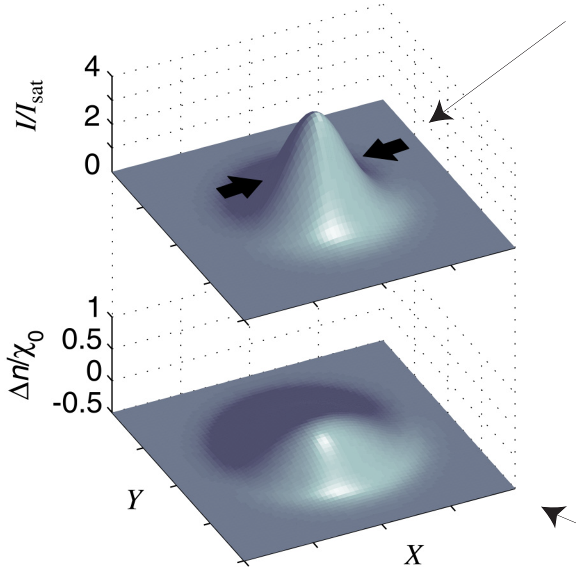
Joe Davis, MIT

Interest in Transverse Effects in NLO

- **Reduction of laser beam filamentation**
- **Generation of quantum states of light**
- **Fundamental interest in nonlinear optical pattern formation**

Spontaneous Pattern Formation in Sodium Vapor

A sodium vapor may be thought of as a medium composed of two-level atoms. Light whose frequency is near the atomic transition frequency experiences a refractive index n which depends strongly on the intensity I:



beam will narrow due to lens-like refractive index profile

induced refractive index is like a positive lens

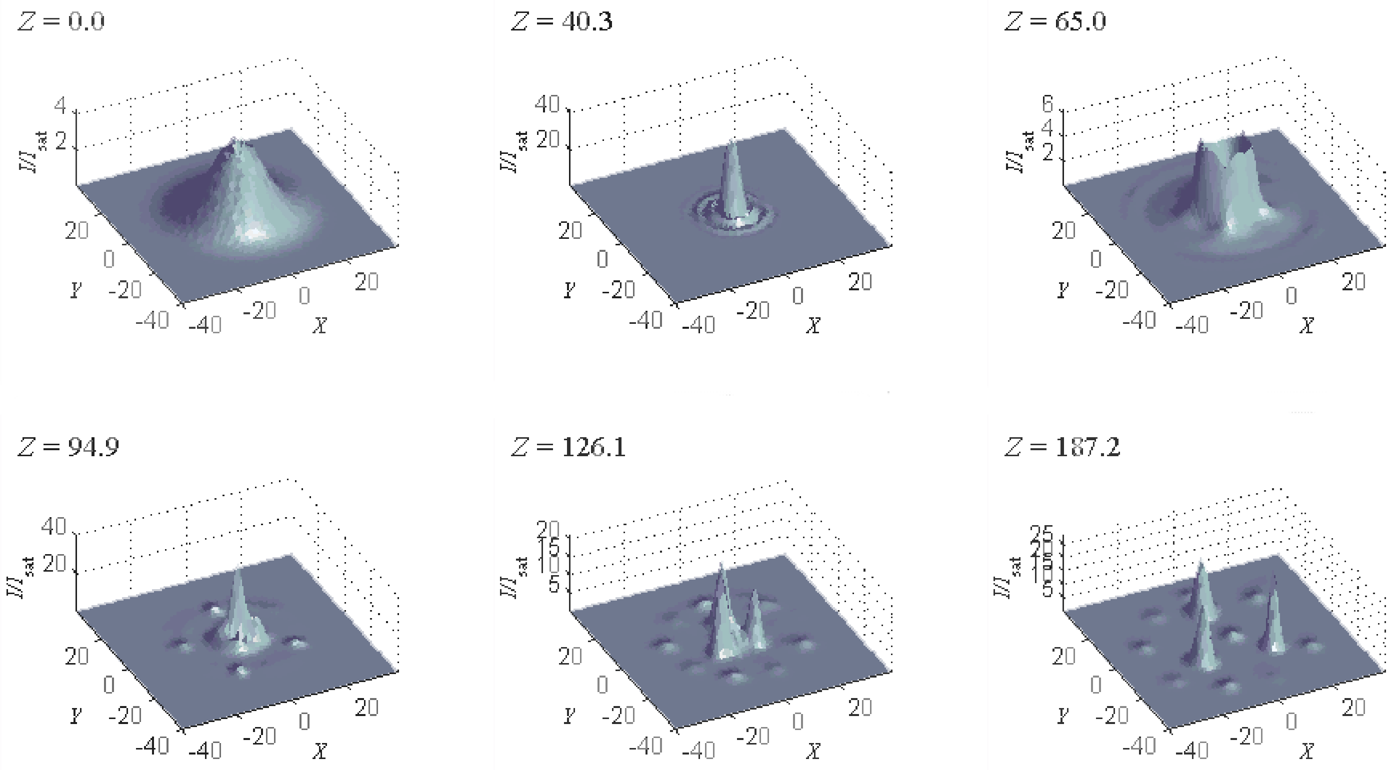
$$n = 1 + \quad n$$

$$1 + \frac{1}{2}\,\frac{0}{1 + I/I_{sat}}$$

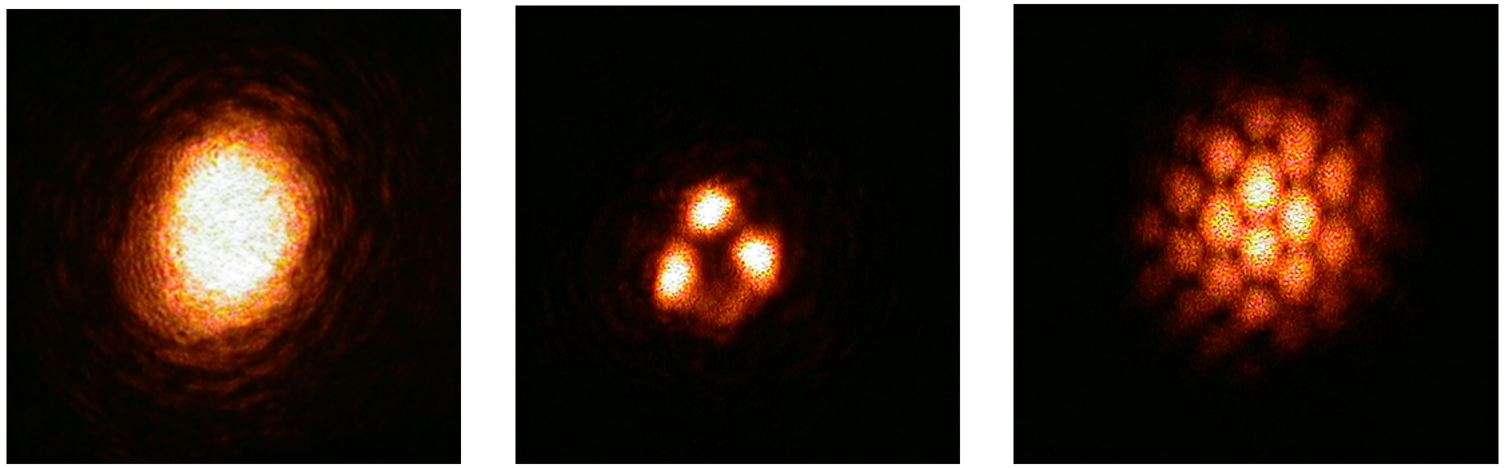
Since light refracts in the direction of increasing index, in a medium with negative saturable nonlinearity it refracts toward regions of higher intensity. This causes smooth beams to narrow or self-focus. But it also tends to destabilize a beam as small amplitude fluctuations grow due to local self-focusing. Thus beams with even small amplitude noise can spontaneously split into two or more separate beams.

**For sodium at 200°C, $_0$ -0.05 and I_{sat} 6 mW/cm^2*

A simulation of spontaneous break-up into 3 stable beams:



Experimental observation of spontaneous break-up resulting in a striking far-field pattern:



beam entering sodium

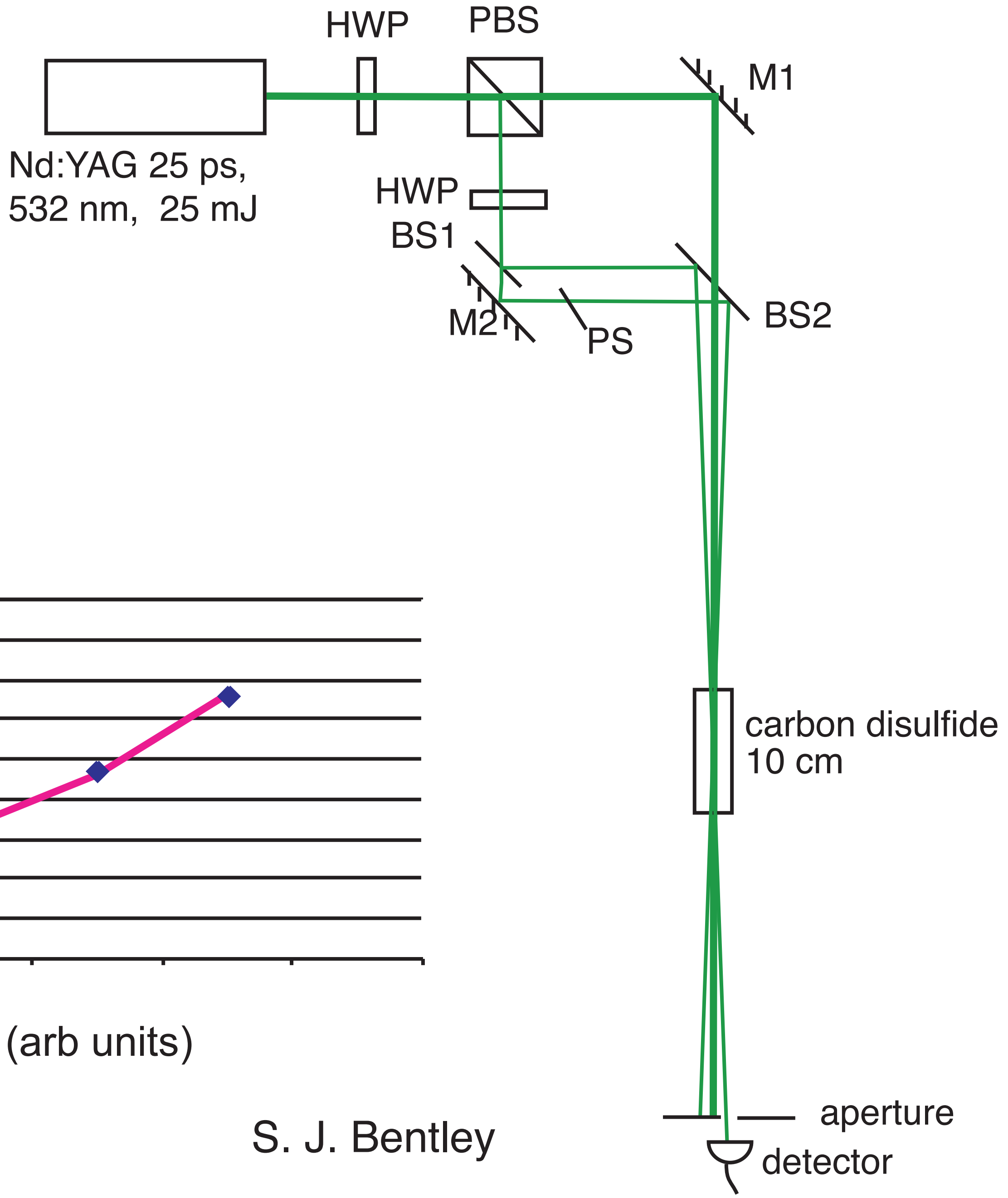
beam leaving sodium

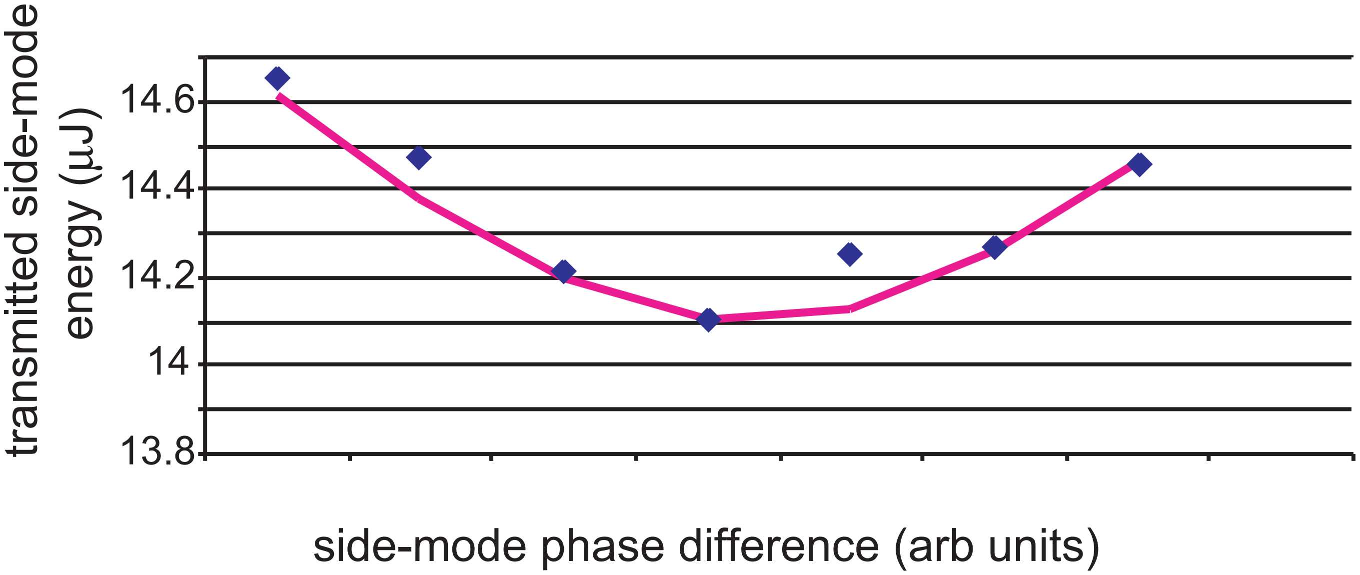
far-field pattern

Pictures taken by R. Bennink, S. Lukishova, and V. Wong.

Preventing Laser-Beam Filamentation

- Use phase to control forward FWM gain
- Control of (laser-beam) decoherence





S. J. Bentley